

Magnetization of a D.C. biased quantum dot

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Using a quantum generalization of the Onsager principle of microscopic reversibility, the magnetization of a system in a non-equilibrium steady state quantum dot is formulated as a response of the interaction energy to an external field. This formulation permits a direct and compact computation of the steady-state magnetization of a non-equilibrium quantum dot as a differential of the interaction energy. Unlike the direct computation of the magnetization using perturbative Keldysh methods, this approach does not require the use of a point splitting procedure. Our results nevertheless support earlier calculations made in the limit of zero field, and they support the survival of strong coupling to arbitrarily large voltages, both at zero field, and under the conditions where the chemical potential difference eV becomes equal to the spin-flip energy in a field $eV = g\mu_B B$.

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I. INTRODUCTION

Quantum dots¹ offer a unique opportunity to study the behavior of a simple, yet non-trivial strongly correlated electron system out of equilibrium.²⁻¹³ In the Coulomb blockade regime, quantum dots with odd numbers of electrons develop a magnetic moment that interacts non-perturbatively with the leads at low temperatures. The unbiased quantum dot provides a classic example of strongly correlated electron physics: at low temperatures, the residual interaction of the dot spin with electrons in the leads gives rise to a Kondo effect, whereby the formation of a many-body resonance at the Fermi energy drives the conductance in the Coulomb blockade regime back up to the unitary limit. The response of the Kondo effect to a D.C. bias is a matter of some interest, and provides us with a chance to examine how a non-trivial strongly correlated electron system responds in a non-equilibrium setting.

One of the key properties of the Kondo effect in equilibrium is the presence of a “running coupling constant”¹⁴ whereby the antiferromagnetic coupling between the dot spin and the leads $g(\Lambda)$ grows progressively as the energy scale Λ is reduced. The Kondo effect exhibits the phenomenon of confinement. At high energies, the local moment is asymptotically free, weakly interacting with the surrounding environment, but at low energies the spin of the dot is “confined”: screened by the lead electron. A single scale, the “Kondo temperature” T_K , governs the low temperature properties; $T_K = D\sqrt{g}e^{-1/2g}$, where g is the ‘bare coupling’ between the spin of the dot and the leads and $D \gg T_K$ is the electron bandwidth. Thus, for example, the magnetization at temperature T and magnetic field B is a universal function $M = m(T/T_K, B/T_K)$, where M has a perturbative “weak coupling” expansion in g only when $T, B \gg T_K$. (See Fig. 1(a).)

What happens to the Kondo effect when a current flows through a D.C. biased quantum dot? Some have suggested that the passage of electrons through the quantum dot will “decohere” the physics of the Kondo effect^{6,15-17}, dephasing the coherent spin-flip processes necessary for the Kondo screening process. The rate at which electrons pass through the quantum dot is given by I/e , where I is the current through the dot. The formation of a Kondo resonance requires that quantum processes are coherent on a time scale $\sim \hbar/T_K$. Suppose that quantum processes on time-scales in excess of $\tau_c \sim e/I$ are dephased by the passage of electrons, then when $\tau_c \lesssim \hbar/T_K$, or when

$$I \gtrsim \frac{eT_K}{\hbar}$$

the coherent formation of a Kondo resonance is expected to break-down. This would imply that the Kondo effect will break down at voltages in excess of the Kondo temperature. In such a picture, the magnetization of the quantum dot

$$M = M(T/T_K, H/T_K, eV/T_K)$$

should become perturbative in the coupling to the leads at arbitrarily low temperatures once $eV \gg T_K$.

An alternative picture of the quantum dot argues that the Kondo effect is modified, but not dephased by the passage of electrons through the dot^{18,19}. According to this picture, although interlead Kondo processes are cut-off by a finite voltage, intralead Kondo renormalization effects continue unabated in each lead until the system enters a non-equilibrium strong-coupling regime at a renormalized temperature $T_K^*(V)$. If true, the DC biased quantum dot should display a Curie like susceptibility characteristic of a decoupled local moment even in the presence of a current, until the temperature approaches

the renormalized Kondo temperature $T_K^*(V)$.

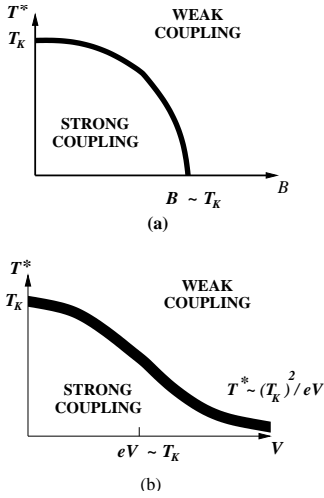


FIG. 1. (a) Field dependence of the crossover temperature $T_K^*(B)$ separating weak- and strong-coupling regimes of the equilibrium Kondo model. At absolute zero, for fields larger than the Kondo temperature the quantum dot re-enters weak coupling. (b) Voltage dependence of the crossover temperature $T_K^*(V)$, from earlier results of Coleman, Hooley and Parcollet¹⁹. Note that T_K^* goes to zero only in the limit $V \rightarrow \infty$, i.e. for low enough temperatures, the model reaches a strong coupling state irrespective of the magnitude of the voltage.

Such a possibility rests on the observation that the Kondo screening process partially delocalizes the spin of the quantum dot into the leads to form a kind of “meso-spin”

$$\vec{S}^*(\Lambda) = U(\Lambda)\vec{S}U^\dagger(\Lambda),$$

whose composite structure depends on the scale on which it is observed. Here, the idea is that same unitary transformation $U(\Lambda)$ which integrates out the high energy electrons also admixes and hence delocalizes the bare dot spin \vec{S} into the leads. (Fig. 2.) In this picture, the rapid fluctuations of the bare quantum dot spin \vec{S} at a rate comparable with τ_c^{-1} , are merely internal redistributions of spin within the scale dependent meso-spin,

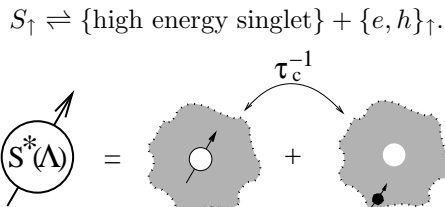


FIG. 2. Renormalized “meso-spin” which forms around a magnetic ion as high-energy electrons are systematically integrated out of the spectrum. The original spin of the bare local moment is partially delocalized into the conduction sea. Fluctuations of the bare moment at a rate τ_c^{-1} lead to an internal redistribution of spin within the meso-spin, but do not cause the composite object to flip.

If this picture is correct, the magnetization of the DC biased quantum dot is expected to remain perturbatively close to a Curie law at large voltages down to a renormalized Kondo temperature $T_K^*(V)$, where a new kind of non-equilibrium Kondo effect will take place.

These discussions suggest that a careful computation of the quantum dot magnetization may provide a key to understanding whether a Kondo effect takes place in the quantum dot at a large voltage bias. In this paper, we present an extended discussion of earlier perturbative calculations of the quantum dot magnetization by Coleman, Hooley and Parcollet¹⁹ that suggest that strong coupling physics extends to arbitrarily high voltages in the D.C. biased Kondo model (Fig. 1 (b)). Two of the three original authors of that work, Parcollet and Hooley²⁰ have recently argued that the key results are incorrect, claiming that the perturbative expansion of the magnetization breaks down at zeroth order in the coupling constants when a current flows through the quantum dot. Part of the reason for doubts about the earlier work stemmed from the necessity of introducing a “point-splitting” procedure to control divergences in the perturbation theory associated with the conservation of the total magnetization. In this paper we present new arguments in favor of the original conclusions of Coleman, Hooley and Parcollet¹⁹. Our arguments avoid some of the earlier difficulties associated with point-splitting by taking advantage of a quantum mechanical extension of Onsager’s principle of microscopic reversibility^{21,22} to steady-state non-equilibrium quantum mechanics.

II. MODEL AND KEY RESULTS

Under conditions where the quantum dot has almost integral valence, interactions between the local moment on the dot and its leads can be described by a Kondo model, of the following form:

$$H = \sum_{m\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} c_{m\mathbf{k}\sigma}^\dagger c_{m\mathbf{k}\sigma} + \mathcal{H}_I - B M_{TOT},$$

$$\mathcal{H}_I = J \psi_\alpha^\dagger(0) \vec{\sigma}_{\alpha\beta} \psi_\beta \cdot \vec{S}. \quad (1)$$

Here, $c_{\alpha\mathbf{k}\sigma}^\dagger$ creates an electron in lead $m \in \{L, R\}$ with momentum \mathbf{k} and spin σ , and J is a positive (antiferromagnetic) Kondo coupling constant derived from virtual charge fluctuations on and off the quantum dot. For simplicity the spectrum of the leads on either side of the dot is taken to be identical and

$$\psi_\alpha(0) = \sum_{\vec{k}} \alpha_L c_{L\mathbf{k}\alpha} + \alpha_R c_{R\mathbf{k}\alpha} \quad (2)$$

describes a linear combination of electrons in the left and right hand-leads with $\alpha_R^2 + \alpha_L^2 = 1$. In equilibrium the left and right-hand leads are filled to the same chemical potential μ , but out-of-equilibrium, the left and right

hand leads differ by an amount equal to the driving voltage

$$\mu_L - \mu_R = eV. \quad (3)$$

Finally, note that we have included the magnetic field term in the interaction part of the Hamiltonian, where

$$\hat{M}_{TOT} = 2S_z + \hat{M}_{leads}$$

is the total magnetization of the leads plus dot,

$$\hat{M}_{leads} = \sum_{\vec{k}, \lambda \sigma} \left[(n_{\lambda \vec{k} \uparrow} - n_{\lambda \vec{k} \downarrow}) \right]$$

is the magnetization of the leads and $n_{\lambda \mathbf{k} \sigma} = c_{\lambda \mathbf{k} \sigma}^\dagger c_{\lambda \mathbf{k} \sigma}$ are the number operators for electrons in the leads.

The interaction term in this Hamiltonian can be divided up into inter and intra-lead terms, as follows

$$\mathcal{H}_I = H_{\text{refl}} + H_{\text{trans}}, \quad (4)$$

where

$$H_{\text{refl}} = J_R \sum_{\mathbf{k}, \mathbf{k}', \sigma, \sigma'} \left(c_{R\mathbf{k}\sigma}^\dagger \vec{\sigma}_{\sigma\sigma'} c_{R\mathbf{k}'\sigma'} \right) \cdot \vec{S} + (R \rightarrow L)$$

$$H_{\text{trans}} = J_{LR} \sum_{\mathbf{k}, \mathbf{k}', \sigma, \sigma'} \left(c_{R\mathbf{k}\sigma}^\dagger \vec{\sigma}_{\sigma\sigma'} c_{L\mathbf{k}'\sigma'} \right) \cdot \vec{S} + (R \leftrightarrow L)$$

describe the spin-flip reflection and transmission of electrons between the leads and the coupling constants

$$J_L = \alpha_L^2 J, \quad J_R = \alpha_R^2 J$$

$$J_{RL} = J_{RL} = \alpha_R \alpha_L J \quad (5)$$

It is sometimes useful to consider the coupling constants J_L , J_R and J_{RL} as independent variables.

In equilibrium, the Kondo model has been solved exhaustively by a number of different methods. Central to our whole understanding of the Kondo model, is the renormalization group. Unfortunately, a complete framework for the renormalization group out-of-equilibrium is not yet available. Much of the current understanding of the Kondo model out-of-equilibrium is based on resummed perturbation theory. A second approach to the problem has been to use a strong-coupling analysis. Each of these approaches is faced with its own particular bias. In a resummed perturbation theory, certain classes of processes are selected, and there is genuine danger that cancellations inherent to the full problem are lost in the process of selecting diagrams. By contrast, the strong coupling approach relies heavily on the linearization of the band, and the assumption that the pertinent band-electron cut-offs in strong coupling are larger than the applied voltage. However, we know from the equilibrium Kondo model, that in some sense, the band-width of the strong-coupling problem is given by the Kondo temperature, so it is not clear how this last assumption can be

reconciled with a voltage larger than the Kondo temperature.

In the face of these uncertainties, we argue that the only unbiased way to address the issue of whether the DC biased Kondo model enters a strong-coupling regime, is to revisit perturbation theory in a non-equilibrium context. There are two obvious physical variables that can be used to characterize the Kondo effect out-of-equilibrium: the current through the dot, and the total magnetization. Perturbative studies of the current¹⁰ through a quantum dot indicate that the expansion

$$I = \frac{e}{\hbar} f(T/T_K, eV/T_K)$$

enters a weak coupling, perturbative regime when $eV \gg T_K$. At first sight, this might be taken as evidence that the Kondo effect returns to weak coupling at large voltage bias. However, the current between two leads at large voltage bias $eV \gg T_K$ involves electrons far from the Fermi surface of each lead, and these electrons would be weakly coupled to the dot spin even under equilibrium conditions.

We shall argue that a more useful quantity to characterize the behavior of a quantum dot out of equilibrium, is the static magnetization. We shall take the magnetization M to refer to the impurity contribution to the *total* magnetization, defined as the difference

$$\hat{M} = \langle \hat{M}_{TOT} \rangle_J - \langle \hat{M}_{leads} \rangle_{J=0}.$$

In the region of weak coupling $\max(T, H)/T_K \gg 1$, the equilibrium magnetization has a perturbative expansion. For low fields, $M(T) = \chi(T)B$, where

$$\chi(T) = \frac{1}{T} \left[1 - 2J\rho - 4(J\rho)^2 \left(\log\left(\frac{De^{(3/4+\gamma)}}{2\pi T}\right) \right) \dots \right] \quad (6)$$

where $\gamma = 0.5772 = -\psi(1)$ is Euler's constant, D is the band-width and ρ is the density of states in each lead. In the same cut-off scheme, the high-field magnetization $B \gg T$ is given by

$$M = 1 \left[1 - J\rho - 2(J\rho)^2 \ln\left(\frac{D}{2B}\right) - \dots \right] \quad (7)$$

The cross-over to strong-coupling is defined by the scale at which the second and first order corrections to the magnetic susceptibility and magnetization become equal. This defines a field scale

$$2B \equiv T_K = D \exp[-1/(2J\rho)] \quad (8)$$

and a temperature scale,

$$T_o = D \frac{e^{\frac{3}{4}+\gamma}}{2\pi} \exp[-1/(2J\rho)] = 0.60 T_K \quad (9)$$

In this paper, we generalize these results to the case of finite DC bias, computing the magnetization to second order in the coupling constant at finite temperature,

field and voltage. In order to do this, we need to define what we mean by the magnetization. We consider the effect of turning on the Kondo coupling constant and computing how the magnetization changes in the steady state, as we slowly increase the coupling constant up to its maximum value. We shall argue that under rather general conditions, the change in the magnetization due to the Kondo interaction can be written in the form

$$\Delta M = -\frac{\partial}{\partial B} \int_0^J \frac{dJ'}{J'} \langle \mathcal{H}_I \rangle_{J', H}$$

where $\langle \mathcal{H}_I \rangle_{J, H}$ is the steady state value of the interaction energy, evaluated at Kondo coupling constant J , in a magnetic field B . The advantage of this formulation, is that it permits a ready generalization of equilibrium methods to a non-equilibrium, steady state situation.

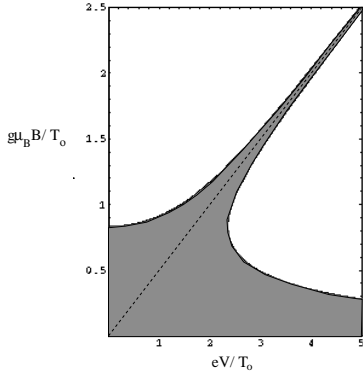


FIG. 3. Showing the region of strong coupling defined by the equality of the first and second order terms in the expansion of the magnetization. Strong coupling persists to arbitrarily high voltage at $B = 0$ and for $g\mu_B B \sim eV$.

The main result of our calculation is succinctly captured by the zero temperature magnetization, which for the symmetric case $J_R = J_L = J_{RL} = J/2$, takes the form

$$M = \left[1 - J\rho - (J\rho)^2 \ln \left(\frac{D^2}{2B\sqrt{(eV)^2 - (2B)^2}} \right) \dots \right] \quad (10)$$

This expression is perturbative so long as

$$2B\sqrt{(eV)^2 - (2B)^2} \gtrsim (T_K)^2 \quad (11)$$

which defines the region shown in Fig. 2.

As can be seen from Fig. 2., large voltage identifies two regions of strong-coupling:

- Zero field Kondo effect $2B < (T_K)^2/eV$, corresponding to a strong coupling region associated with the intra-lead Kondo processes that are not

cut-off by a finite voltage. Earlier work^{18,19} suggested the corresponding fixed point is a two-channel Kondo model where

$$H_I^* \sim [J_R^* \psi_R^\dagger \vec{\sigma} \psi_L + J_L^* \psi_L^\dagger \vec{\sigma} \psi_R] \cdot \vec{S}$$

More recent work by Rosch et al¹⁶ and Schiller et al¹⁷ has suggested that the entry into the strong-coupling two-channel regime is ultimately cut-off by decoherence effects. We shall return to this point in the discussion.

- Finite field Kondo effect. This occurs around $eV \sim 2B \equiv g\mu_B B$, $|eV - 2B| < T_K^4/(eV)^3$, and also extends up to arbitrarily large voltage. Here the Kondo effect involves the degeneracy between a spin flip of the quantum dot and the transfer of an electron between the fermi surface of the two leads. The effective Kondo model for this situation presumably resembles a one-channel Kondo model at zero voltage bias, where the down electrons on high voltage lead and the “up” electrons on the low voltage lead can scatter elastically between leads by exchanging spin with the local moment. We expect that the physics at energy scales lower than $\Lambda \sim \max(g\mu_B B, eV)$, will involve only these electrons, and the effective model will be a one channel Kondo model of the form

$$H_I^* \sim J^* \vec{S} \cdot (\psi_{R\uparrow}^\dagger, \psi_{L\downarrow}^\dagger) \vec{\sigma} \begin{pmatrix} \psi_{R\uparrow} \\ \psi_{L\downarrow} \end{pmatrix} \quad (12)$$

where the spin \vec{S} sits in a zero field, and the chemical potential difference between the $L \downarrow$ and $R \uparrow$ electrons is $\Delta\mu^* = \Delta\mu - g\mu_B B$. In this situation, we expect the spin on the quantum dot to be perfectly quenched at low temperatures. Small changes in the voltage will affect magnetization, however, without a mechanism for the transfer of electrons from one lead to another, this finite field Kondo effect will not, it appears, lead to a unitary differential conductance.

At finite temperatures, our general result takes the form

$$M = (1 - \bar{J}) \tanh(B[1 - \bar{J}]/T) - 2[(\bar{J}_R)^2 + (\bar{J}_L)^2] \frac{\partial}{\partial B} \left[M_o B \left(\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, 0\right) \right) \right] - 4(\bar{J}_{RL})^2 \frac{\partial}{\partial B} \left[M_o B \left(\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, \frac{V}{T}\right) \right) \right] + O(\bar{J}^3) \quad (13)$$

where $\bar{J}_\alpha = J_\alpha \rho$, $\bar{J} = (\bar{J}_R + \bar{J}_L)$ and

$$\phi(b, v) = \text{Re} \int dx \frac{1}{(2 \cosh(\frac{x}{2}))^2} \times \frac{1}{4} \sum_{\sigma, \gamma=\pm 1} 2\pi i \sigma \ln \tilde{\Gamma}(x + \sigma b + \gamma v)$$

$$\tilde{\Gamma}(x) = \Gamma\left(\frac{1}{2} + \frac{x}{2\pi i}\right)$$

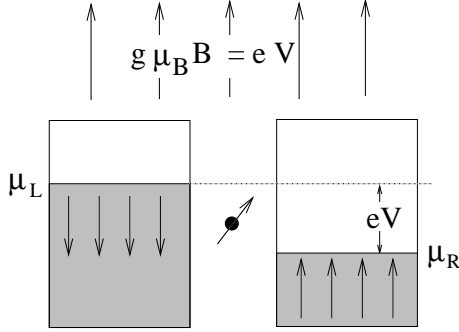


FIG. 4. Illustrating the Kondo effect at a finite field $g\mu_B B = eV_0$. At absolute zero, we expect the ground-state to be a singlet and small departures in the voltage from V_0 will then lead to a magnetic polarization of the local moment.

In the zero field, finite temperature limit, this general result reverts to the form $M = \chi B$, where

$$\chi_i = \frac{1}{T} \left\{ 1 - 2(\bar{J}_R + \bar{J}_L) - 4(\bar{J}_L^2 + \bar{J}_R^2) \ln \left(\frac{De^{\frac{3}{4} + \gamma}}{2\pi T} \right) - 8|\bar{J}_{LR}|^2 \left[\ln \left(\frac{De^{\frac{3}{4} + \gamma}}{2\pi T} \right) - \Phi \left(\frac{V}{T} \right) \right] \right\}. \quad (14)$$

where $\gamma = 0.5772\dots = -\psi(1)$ is the Euler constant. The crossover function $\phi(x)$, in terms of digamma functions $\psi(z)$, is

$$\Phi(x) = \text{Re} \int_{-\infty}^{\infty} \frac{dy}{4 \cosh^2(\frac{y}{2})} \left[\tilde{\psi}(y+x) - \tilde{\psi}(y) \right], \quad (15)$$

where $\tilde{\psi}(x) = \psi(\frac{1}{2} + i\frac{x}{2\pi})$.

The second-order terms in (14) describe the leading logarithmic enhancement of the Kondo coupling. Terms of order J_{LR}^2 involve inter-lead processes and, as expected, the logarithmic divergence in these terms is cut by the voltage. (To see this, note that $-\phi(V/T) \sim -\ln(V/T)$ for $V \gg T$ which cancels the logarithmic temperature divergence.) By contrast, the intra-lead terms of order J_R^2 and J_L^2 are completely unaffected by the voltage V , which guarantees that the leading logarithmic divergence survives at *arbitrarily high voltage*. This is easily seen in the large- V form of the susceptibility,

$$\chi_i = \frac{1}{T} \left\{ 1 - 2(\bar{J}_R + \bar{J}_L) - 4(\bar{J}_L^2 + \bar{J}_R^2) \ln \left(\frac{De^{1+\gamma}}{2\pi T} \right) - 8|\bar{J}_{LR}|^2 \ln \left(\frac{D}{V} \right) \right\}, \quad V \gg T. \quad (17)$$

The survival of the leading logarithms in the susceptibility at arbitrarily high voltage is consistent with the zero-temperature, finite field results, and at least at this

order in perturbation theory, is a signature that the *intra*-lead Kondo effect continues unabated at temperatures or fields smaller than the voltage.

The corresponding boundary where weak-coupling perturbation theory breaks down is shown in Fig. 4.

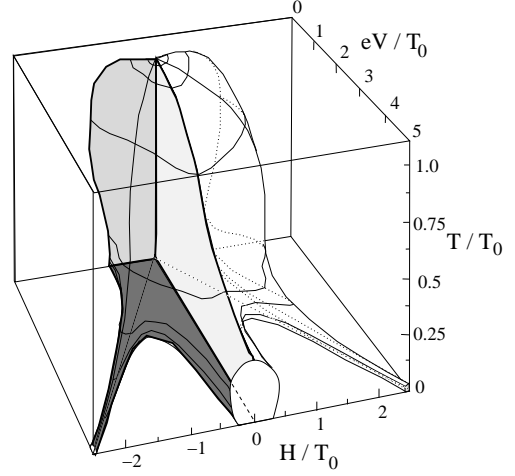


Fig. 4. Illustrating region of strong coupling for finite field, temperature and voltage.

III. MAGNETIZATION AS A RESPONSE FUNCTION

Since we wish to consider the case where the voltage V_{sd} takes an arbitrarily large value, linear response methods are not appropriate; instead, we must resort to the Schwinger-Keldysh formalism for non-equilibrium field theory. The SK method²³ divides the Hamiltonian of the system into two parts:

$$H = H_0 + H_I + \lambda(t)A, \quad (18)$$

where, for convenience we have introduced a source term with field $\lambda(t)$ coupled to quantity A . It is assumed that we know the exact Green's functions for the operators under H_0 . The interaction H_I and source term are regarded as perturbations.

In the SK approach, the expectation value of an operator $A(t_o)$ in the interaction representation $A(t_o) = e^{iH_0 t_o} A e^{-iH_0 t_o}$ is

$$\langle A(t_o) \rangle = \langle P \left(A(t_o) e^{-i \int_K dt [H_I + \lambda(t)A(t)]} \right) \rangle \quad (19)$$

where K denotes the two branch Keldysh contour running from $-\infty$ to $+\infty$ back to $t = -\infty$, (Fig. 5b) P denotes path ordering along this path and $\langle \dots \rangle = \text{Tr}[\rho_o \dots]$ is average with respect to the density matrix ρ_o (in the absence of the interaction) in the distant past.

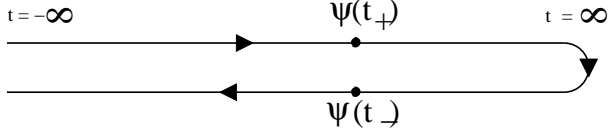


FIG. 5. The Keldysh contour. The closed contour eliminates the need to normalize the path-ordered exponential. Fields are defined on both the upper ($\psi(t_+)$) and lower ($\psi(t_-)$) contour. Along the path, the time t_- is taken to be “later” than the time t_+ .

In the conventional SK approach, the expectation value of some quantity is computed directly, using diagrammatic methods. We may regard this as a direct “measurement” of the quantity in question. In this paper we simplify our computational methods by implementing a new approach which regards expectation values as a response to external fields. The inspiration to go from a “measurement” to a “response” based approach is derived from equilibrium quantum mechanics.

A major difficulty in the Keldysh approach, is that the measurement vertex is much more awkward to use than the “response” vertex. For example, in the Larkin Ovchinnikov basis the response vertex is diagonal whereas the measurement vertex is off-diagonal ($-i\tau_1$). This leads to serious difficulties when dealing with a conserved order parameter, for double poles which appear in the perturbation theory can not be absorbed using integration by parts. The same double poles are easily handled by regarding the expectation value of a quantity as the response to its conjugate field, because the response vertex is proportional to the unit Keldysh matrix, and between two bare Green’s functions it satisfies the relation

$$\mathcal{G}\mathcal{G} = -\partial_\omega \mathcal{G}.$$

This permits us to eliminate double poles in the perturbation theory using integration by parts. It is also extremely useful in the development of Ward identities.

In equilibrium thermodynamics, we first calculate the Free energy, and then we take the derivative to compute the magnetization

$$M = -\frac{\partial F}{\partial B}$$

Can an analogous approach be used out-of-equilibrium?

We now make a digression on the nature of the non-equilibrium steady state. One of the key features of thermal equilibrium, is the notion that the equilibrium steady state does not depend on its past history. In particular, if we turn on some interaction of strength g , $H_I = gh_I$, where h_I is a dimensionless interaction operator, and we couple internal degrees of freedom \hat{A}_i ($i = 1, n$) to corresponding external fields λ_i , then the amount of work done on the system

$$\Delta W = \int_P \langle h_I(t) \rangle dg(t) + \langle A_i(t) \rangle d\lambda_i,$$

where we have used the summation convention on i , does not depend on the detailed path P over which the coupling constants (g, λ_i) are adiabatically incremented to their final value. This implies that the “curl” defined by the following functional derivatives must vanish

$$\begin{aligned} \frac{\delta \langle h_I(t) \rangle}{\delta \lambda_i(t')} - \frac{\delta \langle A_i(t) \rangle}{\delta g(t')} &= 0 \\ \frac{\delta \langle A_j(t) \rangle}{\delta \lambda_i(t')} - \frac{\delta \langle A_i(t) \rangle}{\delta \lambda_j} &= 0. \end{aligned} \quad (20)$$

From linear response theory, we can relate these functional derivatives to the corresponding response functions,

$$\begin{aligned} \frac{\delta \langle h_I(t) \rangle}{\delta \lambda_i(t')} &= -i \langle [h_I(t), A_i(t')] \rangle \theta(t - t') \\ \frac{\delta \langle A_i(t) \rangle}{\delta g(t')} &= -i \langle [A_i(t), h_I(t')] \rangle \theta(t - t') \\ \frac{\delta \langle A_j(t) \rangle}{\delta \lambda_i(t')} &= -i \langle [A_j(t), A_i(t')] \rangle \theta(t - t') \end{aligned} \quad (21)$$

from which it follows that

$$\begin{aligned} -i \langle [h_I(1), A_i(2)] \rangle \theta(1 - 2) &= -i \langle [A_i(1), h_I(2)] \rangle \theta(1 - 2), \\ -i \langle [A_j(1), A_i(2)] \rangle \theta(1 - 2) &= -i \langle [A_i(1), A_j(2)] \rangle \theta(1 - 2). \end{aligned} \quad (22)$$

These relations are the quantum mechanical counterpart of the famous Onsager reciprocity relations^{21,22}. They reflect the microscopic reversibility of the equations of motion and the absence of any “arrow of time” in thermal equilibrium.

We now argue that Onsager’s reciprocity relation will continue to hold for an important sub-class of variables in the non-equilibrium steady state. For our discussion, we consider a quantum dot that is coupled to two very large baths of electrons (“leads”) at different chemical potentials μ_L and μ_R where $\mu_L > \mu_R$. The entire system is isolated. Suppose we connect the quantum dot to the leads at time $t = 0$, then we expect that after some equilibration time τ_1 , which is basically independent of the size of the leads, the system will arrive at a steady state where a current of electrons flows from the left, to the right-hand lead. This steady state will persist for a long time $\tau_2(L)$ until a substantial fraction of the additional electrons on the left lead have flowed into the right lead. Typically, the time $\tau_2(L)$ over which the steady state will persist, will diverge as $L \rightarrow \infty$. The steady state value of some variable A is then given by

$$\langle A \rangle = \lim_{L \rightarrow \infty} \langle A(t) \rangle$$

with the understanding that $\tau_2(L) \gg t \gg \tau_1$.

The steady state involves a persistent current from left to right, and evidently involves an “arrow of time”. Nevertheless, we expect that the nature of the steady state does not depend on the history of how it is arrived at. In particular, suppose we the steady state is arrived by adiabatically turning on an interaction $H_I = gh_I$ between

the leads, and by coupling source terms λ_j to various quantities A_j associated with the dot. Since the system is closed, when we adiabatically change these variables we can define the amount of work done in reaching the steady state

$$\Delta W_{NE} = \int \langle h_I(t) \rangle dg(t) + \langle A_i(t) \rangle d\lambda_i,$$

We emphasize that since the system is completely isolated, the amount of work done in reaching the final state is literally the change in the total energy of the system: a completely well-defined quantity.

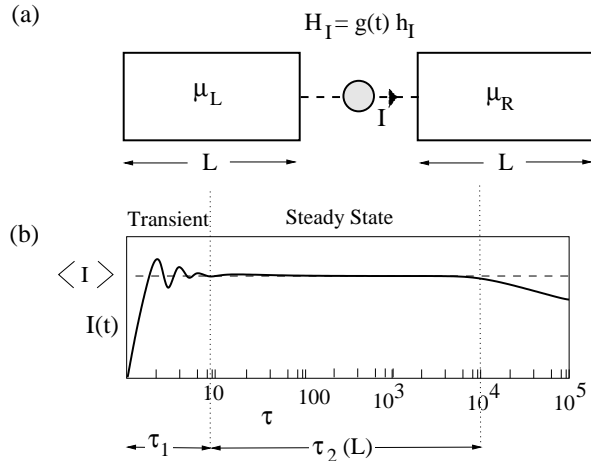


FIG. 6. Illustrating a closed quantum mechanical system in which the non-equilibrium steady state of a quantum dot can be studied. (a) Quantum dot coupled to two isolated leads, initial in thermal equilibrium at two different chemical potentials, via an interaction term $H_I = g(t)h_I$. The coupling constant $g(t)$ is switched on at time $t = 0$. (b) After an initial transient of time τ_1 , the current reaches a steady state value which only depends on the chemical potential difference and the other fields (e.g the magnetic field) applied to the dot. This steady state persists for a large time $\tau_2(L)$ which goes to infinity as the size of the leads is sent to infinity.

If the steady state is independent of the path by which it is arrive at, then it is not unreasonable to expect that there is a sub-class of variables $\{A_i\}$ for which the work ΔW_{NE} is path independent. If this case, that clearly the Onsager relations given above must extend to the non-equilibrium steady state for this subclass of variables. Of course we do not expect the reciprocity relation to extend to *all* variables, as it does in thermal equilibrium, because this would mean that the arrow of time is completely invisible. However, the idea that there exists a subclass of “conforming variables” whose correlation and reponse functions are completely symmetric in time, if true, could be invaluable for studying the non-equilibrium steady state.

This reasoning has motivated us to recently propose a “Quantum Reciprocity Conjecture”²⁶ :

In the non equilibrium steady state, the set of quantum mechanical observables contains a non-trivial subset \mathcal{P} of “protected” quantum observables $\mathcal{P} = \{a_1, a_2, \dots, a_n\}$ whose correlation functions in the steady state are insensitive to the arrow of time, and which consequently satisfy a quantum mechanical analog of the Onsager reciprocity relations

$$\langle [a(1), b(2)] \rangle = \langle [b(1), a(2)] \rangle, \quad (a, b \in \mathcal{P}).$$

Consider the retarded and advanced Green functions between protected variables,

$$G_{ab}^{(R,A)} = \mp i \langle [a(1), b(2)] \rangle \theta_{\pm}(t_2 - t_1) \quad (23)$$

where $\theta_{\pm}(t) = \theta(\pm t)$. Since a and b are hermitian, these are real functions ($G_{ab}^{(R,A)}(t) = [G_{ba}^{(R,A)}(t)]^*$). The conjectured Onsager relations mean that in the steady state, they also satisfy

$$\begin{aligned} G_{ab}^R(t_2 - t_1) &= G_{ab}^A(t_1 - t_2), \\ G_{ab}^{(R,A)}(t_2 - t_1) &= G_{ba}^{(R,A)}(t_2 - t_1), \end{aligned} \quad (24)$$

where the order of the subscripts and time variables is important. If we write $G^R = G^{R*}$ in the first relation, and then Fourier transform, we obtain the more familiar result

$$G_{ab}^A(\omega) = G_{ab}^R(\omega)^*$$

which means that the retarded and advanced Green functions of protected variables share the same spectral decomposition

$$G_{ab}^{(R,A)}(\omega) = \int \frac{dE}{\pi} \frac{1}{\omega - E \pm i\delta} A_{ab}(E)$$

where $A_{ab}(E) = \pm \text{Im}[G_{ab}^{(A,R)}(E)]$.

Onsager used his reciprocity relations to propose that an effective “Free energy” might exist in a non-equilibrium steady state^{21,22}, an idea which has its origin in early works by Lord Rayleigh²⁴. Recently, Derrida, Lebowitz and Speer have shown that this idea holds rigorously in certain simplified non-equilibrium classical models²⁷. Provided that the set of protected quantum variables includes the interaction h_I , then it must follow that one can define an effective Free energy from the virtual work done in reaching the steady state

$$\begin{aligned} \Delta F[g, \{\lambda_j\}] &= \int \langle h_I \rangle dg + \langle A_i \rangle d\lambda_i \\ &= \int \frac{\langle H_I \rangle}{g} dg + \langle A_i \rangle d\lambda_i. \end{aligned} \quad (25)$$

We have tested this conjecture on a simple, non-interacting resonant level model²⁶, where the hybridization with the leads $H_I = gh_I$, the occupancy, magnetization and spin current through the dot are found to

satisfy mutual Onsager reciprocity relations in the presence of a finite steady state charge current. The current operator does not lie within the class of protected variables, nor indeed does any operator which changes the balance of coupling between the leads, or with a separate heat bath. Nevertheless, the existence of a finite subclass of protected variables has been confirmed in this non-interacting example.

We now apply the above arguments on the assumption that the interactions inside the quantum dot are also protected variables. If this is the case, then interactions can be adiabatically turned on inside the quantum dot, whilst preserving the reciprocity relations. This assumption enables us to extend the classic equilibrium expression for the change in magnetization as a consequence of coupling to the leads,

$$\Delta M = -\frac{\partial}{\partial B} \int \frac{dg}{g} \langle H_I \rangle_g^{(Eqn)}$$

to the non-equilibrium situation

$$\Delta \mathcal{M} = -\frac{\partial}{\partial B} \int \frac{dg}{g} \langle H_I \rangle_g^{(NE)},$$

This “energetically defined” magnetization quantity measures the response of the interaction energy to the external field in a the non-equilibrium steady state and it is an interesting quantity in its own right. We shall now show that this quantity is directly related to the change in the non-equilibrium magnetization due to interactions,

$$\Delta M_{NE} = \Delta \mathcal{M}.$$

provided Quantum Reciprocity holds between the interaction and the magnetization.

Consider a non-equilibrium system in the steady state. To be careful, let us focus on some variable in the Hamiltonian, call it A , which is conjugate to variable $\lambda(t)$, so that the Hamiltonian has the form $H = H_o + H_I + \lambda(t)A(t)$, where we implicitly assume that the interaction $H_I = gh_I$ has strength g , which we ultimately set to unity. Imagine turning on the interaction, the change in the A is then given by

$$\frac{\partial A(t_o)}{\partial g} = -i \frac{1}{g} \int_{-\infty}^{t_o} \langle [A(t_o), H_I(t')] \rangle dt'$$

where the response function is to be evaluated at non-zero g . If quantum reciprocity holds,

$$-i \langle [A(t_o), H_I(t')] \rangle = -i \langle [H_I(t_o), A(t')] \rangle,$$

so that we can now put

$$\frac{\partial A(t_o)}{\partial g} = -i \frac{1}{g} \int_{-\infty}^{t_o} \langle [H_I(t_o), A(t')] \rangle dt' \quad (26)$$

But we can simplify this expression by writing

$$-i \langle [H_I(t_o), A(t')] \rangle \theta(t_o - t') = \frac{\delta \langle H_I(t_o) \rangle}{\delta \lambda(t')}$$

so that

$$\begin{aligned} \frac{\partial A(t_o)}{\partial g} &= -\frac{1}{g} \int_{-\infty}^{t_o} dt' \frac{\delta \langle H_I(t_o) \rangle}{\delta \lambda(t')} \\ &= \frac{1}{g} \frac{\partial \langle H_I(t_o) \rangle}{\partial \lambda}. \end{aligned} \quad (27)$$

Finally, integrating over g , we have

$$\Delta A(t_o) = \frac{\partial}{\partial \lambda} \int_0^1 \frac{dg'}{g'} \langle H_I(t_o) \rangle_{g'}$$

We will apply this expression to the magnetization in the non-equilibrium Kondo model, making the replacement $A \rightarrow M$ and $\lambda \rightarrow -B$ and $g \rightarrow J$ to obtain

$$\Delta M = -\frac{\partial}{\partial B} \int \frac{dJ}{J} \langle H_I \rangle^{(NE)}.$$

This expression deserves some discussion. In deriving it, we have assumed that the magnetization can relax to its new steady state value : a very delicate assumption!

The magnetization is actually a conserved quantity which commutes with the full Hamiltonian $[M, H] = 0$, and so strictly speaking, by coupling the dot to the leads, we fail to relax the total magnetization. To avoid this paradox, we must surreptitiously introduce a regulated magnetization operator whose matrix elements lie between states which are not degenerate, as in the case of a conserved operator, but whose energies are split by an energy scale s . Such an operator will consequently relax on a time scale $\tau_r = \hbar/s$. It is important that this time-scale is faster than the adiabatic time scale $\tau_a \sim \hbar/\delta$ at which the interaction is switched on, where δ is the regulator used inside the Green functions, i.e.

$$\tau_r = \hbar/s < \hbar/\delta = \tau_a.$$

This is done in practice by weakly connecting the leads and the local moment to a heat bath, e.g. by considering the local moment to weakly coupled to a spin chain. Formally, at the end of the calculation, we take $\delta \rightarrow 0$ at fixed s , then let s go to zero:

$$M = \lim_{s \rightarrow 0} \lim_{\delta \rightarrow 0} \langle M \rangle_{s, \delta}$$

Formally, a precisely similar regulation is required in equilibrium. For example, when we consider the Pauli spin susceptibility, which is computed by taking the spin susceptibility at a finite q which is ultimately set to zero.

$$\chi_{Pauli} = \lim_{q \rightarrow 0} \lim_{\omega \rightarrow 0} \chi(\vec{q}, \omega)$$

Fortunately, we need only consider this process in the abstract, because the computation of the interaction energy in a field does not present one with any divergent singularities. Difficulties in earlier calculations associated with point-splitting are completely eliminated.

IV. DEFINITION OF GREEN FUNCTIONS

Let us denote by t_+ a time lying on the upper branch of the contour, and by t_- a time lying on the lower branch. Then we may formally define four Green's functions:

$$\begin{aligned} G^{++}(t, t') &\equiv -i\langle T\psi(t_+)\psi^\dagger(t'_+)\rangle; \\ G^{+-}(t, t') &\equiv i\langle \psi^\dagger(t'_+)\psi(t_-)\rangle; \\ G^{-+}(t, t') &\equiv -i\langle \psi(t_-)\psi^\dagger(t'_+)\rangle; \\ G^{--}(t, t') &\equiv -i\langle \tilde{T}\psi(t_-)\psi^\dagger(t'_-)\rangle. \end{aligned}$$

Here, T is the usual time ordering operator, while \tilde{T} is the anti-time-ordering operator; we consider the operators ψ to be fermionic. The Green's functions in (28) are not all independent. In fact, there are only two independent Green's functions here. This is most easily seen by employing a transformation into the so-called ‘‘Larkin-Ovchinnikov’’ basis²⁵.

Let us define the matrix \mathbf{G}' , whose entries are the four Green's functions of (28):

$$\mathbf{G}' \equiv \begin{pmatrix} G^{++} & G^{+-} \\ G_{-+} & G_{--} \end{pmatrix}. \quad (28)$$

The transformation to the Larkin-Ovchinnikov basis is defined as

$$\mathbf{G}' \rightarrow \mathbf{G} \equiv \mathbf{U}\mathbf{G}'\mathbf{V}, \quad (29)$$

where the matrices \mathbf{U} and \mathbf{V} are given by

$$\mathbf{U} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}, \quad \mathbf{V} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix}. \quad (30)$$

The elements of this matrix Green-function correspond to the Green-functions of the symmetric and antisymmetric fields

$$\begin{aligned} \psi_S(t) &= \frac{1}{\sqrt{2}} (\psi(t_+) + \psi(t_-)) \\ \psi_A(t) &= \frac{1}{\sqrt{2}} (\psi(t_+) - \psi(t_-)). \end{aligned} \quad (31)$$

Defining

$$\Psi(t) = \begin{pmatrix} \psi_S \\ \psi_A \end{pmatrix}, \quad \bar{\Psi}(t) = \psi^\dagger(t)\tau_1 = (\psi_A^\dagger, \psi_S^\dagger) \quad (32)$$

then

$$\begin{aligned} \mathbf{G}(1, 2) &= -i\langle P\Psi(1)\bar{\Psi}(2) \rangle \\ &= -i\langle P \left[\begin{pmatrix} \psi_S(1) \\ \psi_A(1) \end{pmatrix} \otimes (\psi_A^\dagger(2) \quad \psi_S^\dagger(2)) \right] \rangle \end{aligned} \quad (33)$$

where P denotes ‘‘path-ordering’’ of the fields along the Keldysh contour. It is easily shown that the joint fluctuations in the antisymmetric fields identically vanish, (Appendix A)

$$G_Y(1, 2) = -i\langle P\psi_A(1)\psi_A^\dagger(2) \rangle = 0, \quad (34)$$

and that the remaining matrix elements can be written as follows

$$\mathbf{G} = \begin{pmatrix} G^R & G^K \\ 0 & G^A \end{pmatrix}, \quad (35)$$

where the physical Green's functions are given by

$$\begin{aligned} G^R(t, t') &= -i\theta(t - t') \langle \{ \psi(t), \psi^\dagger(t') \} \rangle; \\ G^A(t, t') &= i\theta(t' - t) \langle \{ \psi(t), \psi^\dagger(t') \} \rangle; \\ G^K(t, t') &= -i \langle [\psi(t), \psi^\dagger(t')] \rangle. \end{aligned} \quad (36)$$

The first two Green's functions encode the dynamics whereas, the third, G^K (the Keldysh Green's function), encodes the evolution of the particle distribution. This, of course, is unnecessary in equilibrium, since the particle distribution is known if the dynamic Green's functions are given. The two indices of the matrix \mathbf{G} will be called ‘‘indices in Keldysh space’’.

Let us now discuss the specific propagators for the quantum dot Kondo model. One technical difficulty is that the Hamiltonian H_I contains, the spin operator \mathbf{S} . To furnish a Wick's theorem, it proves convenient to factorize \mathbf{S} in terms of canonical creation and annihilation operators. This is the ‘‘Abrikosov pseudo-fermion representation’’²⁸

$$\vec{S} = f^\dagger_\alpha \left(\frac{\vec{\sigma}}{2} \right)_{\alpha\beta} f_\beta, \quad (37)$$

where f^\dagger_α creates a pseudo-fermion with spin component α . To provide a faithful representation of the spin 1/2 operator, we must impose the additional constraint $n_f = 1$. This is done very conveniently by using the ‘‘Fedatov-Popov trick’’^{29,30}, in which the f-electron field in the past is equilibrated with a heat bath at a complex chemical potential $\mu = -i\pi T/2$. With this device, the contributions of doubly occupied and empty states exactly cancel. The occupancy of the f-electrons in the distant past is

$$n_{f\sigma} = f(-\sigma B + i\pi T/2) = \frac{1}{e^{-\sigma B/T + i\pi/2} + 1},$$

where $f(x)$ is the Fermi function. The magnetization of a free spin then recovers the Brillouin function

$$M^{(0)}(T, B) = \sum_{s=\pm 1} \sigma n_{fs} = \tanh\left(\frac{B}{T}\right).$$

yielding a Curie susceptibility $\chi^0 = \partial M / \partial B|_{B=0} = \frac{1}{T}$.

The diagrammatic elements corresponding to the bare Green's functions are then

$$\text{---} \xrightarrow[\mathbf{k}, \omega]{\mathbf{m}} \text{---} = \mathbf{g}_m(k, \omega) \quad (38)$$

for the conduction electron in the m -th lead and

$$- \text{---} - \text{---} \blacktriangleright - \text{---} = \mathbf{G}(\omega) \quad (39)$$

for the pseudofermion. Written out explicitly,

$$\begin{aligned} [\mathbf{g}_m(k, \omega)]_{\sigma\sigma'} &\equiv \mathbf{g}_{m\sigma}(k, \omega) \delta_{\sigma\sigma'}, \\ \mathbf{g}_{m\sigma}(k, \omega) &= \begin{pmatrix} g_{m\sigma}^R(k, \omega) & g_{m\sigma}^K(k, \omega) \\ 0 & g_{m\sigma}^A(k, \omega) \end{pmatrix} \\ &= \begin{pmatrix} \frac{1}{\omega - \epsilon_{\mathbf{k}\sigma} + i\delta} & 2\pi i h_m(\epsilon_{\mathbf{k}\sigma}) \delta(\omega - \epsilon_{\mathbf{k}\sigma}) \\ 0 & \frac{1}{\omega - \epsilon_{\mathbf{k}\sigma} - i\delta} \end{pmatrix}, \quad (40) \end{aligned}$$

where $\epsilon_{\mathbf{k}\sigma} = \epsilon_{\mathbf{k}} - \sigma B$ and

$$\begin{aligned} [\mathbf{G}(\omega)]_{\sigma\sigma'} &\equiv \mathbf{G}_\sigma(\omega)\delta_{\sigma\sigma'} \\ \mathbf{G}_\sigma(\omega) &= \begin{pmatrix} G_\sigma^R(\omega) & G_\sigma^K(\omega) \\ 0 & G_\sigma^A(\omega) \end{pmatrix} \\ &= \begin{pmatrix} \frac{1}{\omega - \lambda_\sigma + i\delta} & 2\pi i h_\sigma \delta(\omega - \lambda_\sigma) \\ 0 & \frac{1}{\omega - \lambda_\sigma - i\delta} \end{pmatrix}. \end{aligned} \quad (41)$$

where we have introduced the notation $\lambda_\sigma = -\sigma B$. The functions $h_m(\epsilon) = h(\epsilon - \mu_m)$ and $h_\sigma = h(\lambda_\sigma + \frac{i\pi T}{2})$ give the occupancies of the band and f-electron states, where

$$h(\epsilon) \equiv 2f(\epsilon) - 1 = -\tanh\left(\frac{\epsilon}{2T}\right). \quad (42)$$

The quantity δ is the usual infinitesimal regulator distinguishing the retarded and advanced Green's functions.

V. FEYNMAN RULES

In developing the Feynman rules we need to consider two types of vertices. The “scattering vertex” generated by contractions of terms $H_I^A = H_I(t_+) - H_I(t_-)$ in the expansion of the time ordered exponential is denoted by

$$\begin{aligned}
& \begin{array}{c} \text{Diagram 1: A vertex with four external lines. Top-left: solid line with arrow pointing up-left, labeled } m \text{ and } a\lambda. \text{ Top-right: dashed line with arrow pointing up-right, labeled } \beta\lambda'. \text{ Bottom-left: solid line with arrow pointing down-left, labeled } n. \text{ Bottom-right: dashed line with arrow pointing down-right, labeled } \alpha\gamma. \end{array} \\
& \equiv \begin{array}{c} \text{Diagram 2: A vertex with four external lines. Top-left: solid line with arrow pointing up-left. Top-right: dashed line with arrow pointing up-right. Bottom-left: solid line with arrow pointing down-left. Bottom-right: dashed line with arrow pointing down-right. A shaded triangle is on the top-left edge.} \end{array} + \begin{array}{c} \text{Diagram 3: A vertex with four external lines. Top-left: solid line with arrow pointing up-left. Top-right: dashed line with arrow pointing up-right. Bottom-left: solid line with arrow pointing down-left. Bottom-right: dashed line with arrow pointing down-right. A shaded triangle is on the bottom-right edge.} \end{array} \\
& = \frac{J_{mn}}{2} \vec{\sigma}_{ab} \cdot \vec{\sigma}_{\alpha\beta} \left[\left(\frac{i\mathbf{T}_1}{2} \right)_{\lambda\lambda'} \otimes \mathbf{1}_{\gamma\gamma'} + \mathbf{1}_{\lambda\lambda'} \otimes \left(\frac{i\mathbf{T}_1}{2} \right)_{\gamma\gamma'} \right]. \quad (43)
\end{aligned}$$

(where the factor “ i ” derives from one factor of $-i$ in the time ordered, exponential, and two factors of i transferred from external legs). Here, α, β, a, b refer to the spin indices and $\lambda, \lambda', \gamma, \gamma'$ to the Keldysh indices of the outgoing and incoming propagators. The factors in bold are the matrices in Keldysh space that appear at the vertex. The shaded triangle denotes the vertex to which the Keldysh matrix $\frac{i\tau_1}{2}$ is to be applied. The Keldysh matrix entering into the vertex is actually symmetric under exchange of the Keldysh indices of the incoming, or outgoing fermions,

$$\begin{aligned}
(\tau_1)_{\lambda\lambda'} \otimes \mathbf{1}_{\gamma\gamma'} + \mathbf{1}_{\gamma\gamma'} \otimes (\tau_1)_{\lambda\lambda'} = \\
(\tau_1)_{\lambda\gamma'} \otimes \mathbf{1}_{\gamma\lambda'} + \mathbf{1}_{\gamma\lambda'} \otimes (\tau_1)_{\lambda\gamma'} \quad (44)
\end{aligned}$$

so that at each scattering vertex one has a choice as which pairs of incoming and outgoing fermion lines one applies the Keldysh matrices to.

Diagrammatically,

$$\begin{array}{c} \diagup \diagdown \\ \diagdown \diagup \end{array} + \begin{array}{c} \diagdown \diagup \\ \diagup \diagdown \end{array} \equiv \begin{array}{c} \diagup \diagdown \\ \diagdown \diagup \end{array} + \begin{array}{c} \diagdown \diagup \\ \diagup \diagdown \end{array} \quad (45)$$

The “measurement vertex” generated by contractions with $H_I^S = \frac{1}{2}[H_I(t_+) + H_I(t_-)]$ is denoted by

$$\begin{aligned}
& \text{Diagram 1: A vertex (circle) with incoming lines } m \text{ (solid arrow) and } n \text{ (solid arrow), and outgoing lines } \beta\lambda' \text{ (dashed arrow) and } \alpha\gamma \text{ (dashed arrow). Labels } a\lambda \text{ and } b\gamma' \text{ are on the incoming lines.} \\
& \equiv \text{Diagram 2: A shaded box with a central dot, with incoming lines } m \text{ (solid arrow) and } n \text{ (solid arrow), and outgoing lines } \beta\lambda' \text{ (dashed arrow) and } \alpha\gamma \text{ (dashed arrow).} \\
& = \frac{J_{mn}}{2} \vec{\sigma}_{ab} \cdot \vec{\sigma}_{\alpha\beta} \left[\left(\frac{i\tau_1}{2} \right)_{\lambda\lambda'} \otimes \left(\frac{i\tau_1}{2} \right)_{\gamma\gamma'} \right], \quad (46)
\end{aligned}$$

(where the factor “ $\epsilon^2 = -1$ ” from the τ_1 matrices is transferred from the contractions of two external propagators.) Although the vertex $(\tau_1)_{\lambda\lambda'} \otimes (\tau_1)_{\gamma\gamma'}$ is not symmetric under interchange of incoming Keldysh indices, one can make it symmetric by adding in the additional “null” term $\mathbf{1}_{\lambda\lambda'} \otimes \mathbf{1}_{\gamma\gamma'}$. The expectation value of this vertex is zero, since it involves the product of two operators that are antisymmetric between the upper and lower Keldysh contour. By introducing this operator, using the identity

$$(\tau_1)_{\lambda\lambda'} \otimes (\tau_1)_{\gamma\gamma'} + \mathbf{1}_{\lambda\lambda'} \otimes \mathbf{1}_{\gamma\gamma'} = (\tau_1)_{\lambda\gamma'} \otimes (\tau_1)_{\gamma\lambda'} + \mathbf{1}_{\lambda\gamma'} \otimes \mathbf{1}_{\gamma\lambda'}$$

and then removing the null operator on the right-hand side, one also has the freedom to interchange the Keldysh indices on the measurement vertex, i.e.

$$\text{Diagram 1} \equiv \text{Diagram 2} \quad (47)$$

Our Feynman rules for the evaluation of the n-th order contribution to $\langle H_I \rangle$ are then

- Construct a connected diagram with one measurement vertex (46) and n scattering vertices (43)
- Associate factors \mathbf{g}_m and $\mathbf{G}(\omega)$ with the conduction and pseudofermion propagators.
- Associate a factor -1 with each conduction electron and pseudo-fermion loop.
- Carry out the trace over spin and Keldysh indices.
- Carry out the summation over internal momenta $\sum_{\mathbf{k}}$.

- Carry out the integration over loop frequencies $\int \frac{d\omega}{2\pi}$. Associate the convergence factor $\cos \omega 0^+ = \frac{1}{2} (e^{i\omega 0^+} + e^{-i\omega 0^+})$ with propagators which form closed loops.
- Associate a symmetry factor $1/n$ with diagrams with permutation symmetry group of dimension n .

VI. SOME USEFUL DIAGRAMS

A. Vanishing Diagrams

In constructing the Feynman diagrams, there are a number of key diagrams which identically vanish. Any loop diagram which does not contain an insertion of the Keldysh matrix $i\tau_1$ automatically vanishes, because it corresponds to the expectation value of a single, or a product of antisymmetric densities. For example, the simple fermion loop corresponds to the equal time expectation value of the difference between fermion number on the upper and lower Keldysh contour

$$\begin{aligned} \text{Diagram} &= -i \langle \bar{f}_\sigma(t) f_\sigma(t) \rangle \\ &= -i \langle n_{f\sigma}(t_+) - n_{f\sigma}(t_-) \rangle = 0 \end{aligned} \quad (48)$$

In the frequency domain this result is obtained as follows

$$\begin{aligned} \text{Diagram} &= - \int \frac{d\omega}{2\pi} \text{Tr}[\mathbf{G}_\sigma(\omega)] \frac{1}{2} (e^{i\omega 0^+} + e^{-i\omega 0^+}) \\ &= - \int \frac{d\omega}{4\pi} [G_\sigma^R(\omega) e^{-i\omega 0^+} + G_\sigma^A(\omega) e^{i\omega 0^+}] \\ &= -\frac{1}{\pi} (-i\pi + i\pi) = 0. \end{aligned} \quad (49)$$

In this diagram, the first convergence factor catches the pole contribution from G_A , whereas the second convergence factor catches an equal yet opposite contribution from G_R . Two other examples of these cancellations are

$$\text{Diagram} = -i \langle n_{mk\sigma}(t_+) - n_{mk\sigma}(t_-) \rangle = 0. \quad (50)$$

and

$$\begin{aligned} \text{Diagram} &= - \int \frac{d\nu}{2\pi} \text{Tr}[\mathbf{G}_\sigma(\omega + \nu) \mathbf{g}_{m\sigma}(k, \nu)] \\ &= 0. \end{aligned} \quad (51)$$

where in this case, the cancellation does not require a convergence factor.

B. Magnetization and Susceptibility

The inclusion of the factor $-i\tau_1/2$ into fermion loops gives the equilibrium expectation value of one particle quantities. For example, the equilibrium magnetization of the local moment is given by

$$\begin{aligned} \text{Diagram} &= -1 \int \frac{d\omega}{2\pi} \text{Tr} \left(\frac{i\tau_1}{2} \sigma_3 \mathbf{G}(\omega) \right) \\ &= \sum_\sigma \sigma \int \frac{d\omega}{4\pi i} [2\pi i h_\sigma \delta(\omega - \lambda_\sigma)] \\ &= \sum_\sigma \frac{\sigma}{2} h(\lambda_\sigma - i\pi T/2) \\ &= M_o(T, B) = \tanh \left(\frac{B}{T} \right). \end{aligned} \quad (52)$$

Using the identity

$$-\frac{\partial}{\partial B} \mathbf{G}(\omega) = \mathbf{G}(\omega) \sigma_3 \mathbf{G}(\omega) \quad (53)$$

we can differentiate the above result to compute the equilibrium finite-field susceptibility of the local moment

$$\begin{aligned} -\frac{\partial}{\partial B} \left[\text{Diagram} \right] &= \sigma_3 \text{Diagram} \sigma_3 \\ &= -1 \int \frac{d\omega}{2\pi} \text{Tr} \left(\frac{i\tau_1}{2} \sigma_3 \mathbf{G}(\omega) \sigma_3 \mathbf{G}(\omega) \right) \\ &= -\frac{\partial M_o}{\partial B} \end{aligned} \quad (54)$$

In a similar fashion the magnetization of the electrons in the m -th lead is given by

$$\begin{aligned} \text{Diagram} &= -1 \sum_k \int \frac{d\omega}{2\pi} \text{Tr} \left(\frac{i\tau_1}{2} \sigma_3 \mathbf{g}_m(k, \omega) \right) \\ &= \sum_{k, \sigma} \frac{\sigma}{2} h_m(\epsilon_{k\sigma}) \\ &= 2B \sum_k \left(-\frac{\partial f_m}{\partial \epsilon_k} \right) = 2\rho B \end{aligned} \quad (55)$$

This is the only case where we have to explicitly consider the effect of the magnetic field on the conduction electron lines. For all conduction electron propagators that do not loop back on themselves, we can carry out the summation over momentum k in advance of forming the Feynman diagram. We shall denote

$$\text{Diagram} = \sum_k \mathbf{g}_m(k, \omega) = \mathbf{g}_m(\omega) \delta_{\sigma\sigma'} \quad (56)$$

where

$$\begin{aligned} \mathbf{g}_m(\omega) &= \begin{pmatrix} g_m^R(\omega) & g_m^K(\omega) \\ 0 & g_m^A(\omega) \end{pmatrix} \\ &= \begin{pmatrix} -i\pi\rho & 2\pi i \rho h_m(\omega) \\ 0 & i\pi\rho \end{pmatrix}, \end{aligned} \quad (57)$$

where we have used the large band-width approximation

$$\sum_k \frac{1}{\omega - \epsilon_{k\sigma} \pm i\delta} = \mp i\pi\rho. \quad (58)$$

In this approximation, the *local* magnetic susceptibility of the conduction electrons is given by

$$\begin{aligned} \sum_{k,k'} \sigma_3 \begin{array}{c} \text{m,k}\omega \\ \text{n,k'}\omega \end{array} \sigma_3 &= - \int \frac{d\omega}{2\pi} \text{Tr} \left[\frac{i\tau_1}{2} \sigma_3 \mathbf{g}_m(\omega) \sigma_3 \mathbf{g}_n(\omega) \right] \\ &= - \frac{\rho}{2} \int d\omega [h_m(\omega) - h_n(\omega)] = 0. \end{aligned} \quad (59)$$

(a form of the Anderson-Clogston compensation theorem.)

C. Kondo Polarization Bubbles

Finally, for our calculations, it proves useful to be able to compute the joint polarization bubbles between the f-electron and the conduction electrons. These bubbles control the renormalization of the interaction between the local moment and the leads.

There are three basic bubbles:

$$\begin{aligned} \omega \leftarrow \begin{array}{c} \text{g} \\ \text{m},\sigma' \end{array} &= \pi_{m\sigma}^R(\omega) \\ \omega \leftarrow \begin{array}{c} \text{g} \\ \text{m},\sigma' \end{array} &= \pi_{m\sigma}^A(\omega) = \pi_{m\sigma}^R(\omega)^* \\ \omega \leftarrow \begin{array}{c} \text{g} \\ \text{m},\sigma' \end{array} &= \pi_{m\sigma}^K(\omega) \end{aligned}$$

Note that the spin index σ on each bubble refers to the spin component of the pseudo-fermion, and that each bubble is independent of the spin component (σ') of the conduction electron propagator. The calculation of the first bubble proceeds as follows

$$\begin{aligned} \omega \leftarrow \begin{array}{c} \text{g} \\ \text{m},\sigma' \end{array} &= - \int \frac{d\nu}{2\pi} \text{Tr} \left[\frac{i\tau_1}{2} \mathbf{G}_\sigma(\omega + \nu) \mathbf{g}_m(\nu) \right] \\ &= \int \frac{d\nu}{4\pi i} \left[G_\sigma^R g_m^K + G_\sigma^K g_m^A \right]_{\omega+\nu,\nu} \\ &= \frac{1}{2} \sum_{k'} \left(\frac{h_m(\epsilon_{k'\sigma'}) - h_\sigma}{\omega + \epsilon_{k'\sigma'} - \lambda_\sigma + i\delta} \right). \end{aligned} \quad (60)$$

In the large large band-width approximation, this becomes

$$\pi_{m\sigma}^R(\omega) = \frac{\rho}{2} \int d\nu \left(\frac{h(\nu - \mu_m) - h_\sigma}{\omega + \nu - \lambda_\sigma + i\delta} \right). \quad (61)$$

Note how the field dependence of the conduction electron propagator has been absorbed in the large band-width limit by a small shift in the conduction electron energies $\epsilon_{k\sigma'} \rightarrow \epsilon_k$. By making use of the identity

$$\frac{1}{2} \int dx \frac{h(x) - h_\sigma}{x - \epsilon + i\Delta} = \psi \left(\frac{1}{2} - \frac{\epsilon - i\Delta}{2\pi iT} \right) - \ln \left(\frac{De^{-\frac{i\pi}{2} h_\sigma}}{2\pi T} \right) \quad (62)$$

where $\Delta > 0$, and $\psi(z) = d \ln \Gamma(z)/dz$ is the digamma function, we obtain

$$\begin{aligned} \pi_\sigma^R(\omega) &= \rho \left[\psi \left(\frac{1}{2} + \frac{\omega - \lambda_\sigma + \mu_m}{2\pi iT} \right) - \ln \left(\frac{De^{-\frac{i\pi}{2} h_\sigma}}{2\pi T} \right) \right], \\ \pi_\sigma^A(\omega) &= \rho \left[\psi \left(\frac{1}{2} - \frac{\omega - \lambda_\sigma + \mu_m}{2\pi iT} \right) - \ln \left(\frac{De^{\frac{i\pi}{2} h_\sigma}}{2\pi T} \right) \right]. \end{aligned} \quad (63)$$

The Keldysh loop is computed as follows

$$\begin{aligned} \omega \leftarrow \begin{array}{c} \text{g} \\ \text{m},\sigma' \end{array} &= - \int \frac{d\nu}{2\pi} \text{Tr} \left[\frac{i\tau_1}{2} \mathbf{G}_\sigma(\omega + \nu) \frac{i\tau_1}{2} \mathbf{g}_m(\nu) \right] \\ &= \int \frac{d\nu}{8\pi} \left[G_\sigma^R g_m^A + G_\sigma^A g_m^R + G_\sigma^K g_m^K \right]_{\omega+\nu,\nu} \\ &= \frac{\pi}{2} \sum_k [1 - h_\sigma h_m(\epsilon_{k\sigma'})] \delta(\lambda_\sigma - \omega - \epsilon_{k\sigma'}). \end{aligned} \quad (64)$$

so that in the large band-width limit

$$\pi_\sigma^K(\omega) = \frac{\pi\rho}{2} [1 - h_\sigma h_m(\lambda_\sigma - \omega)]. \quad (65)$$

VII. DETAILED CALCULATION

We now discuss the evaluation of the interaction energy, up to second-order perturbation theory. The corresponding Feynman diagrams are

$$\begin{aligned} \int_0^J \frac{dJ'}{J'} \langle \mathcal{H}_I \rangle_{J',H} &= \text{Diagram 1} \\ &+ \frac{1}{2} \left[\text{Diagram 2} + \text{Diagram 3} + \text{Diagram 4} \right] \\ &= \Delta F_{(1)} + \Delta F_{(2)} + \Delta F_{(3)} + \Delta F_{(4)} \end{aligned}$$

where an additional factor of 1/2 in the last two terms comes from the coupling constant integration.

In evaluating these diagrams, it is useful to divide the interaction vertex into an “Ising” and “x-y” component, writing

$$\frac{J_{mn}}{2} \vec{\sigma}_{ab} \cdot \vec{\sigma}_{\alpha\beta} = J_{mn} \left[\frac{1}{2} \sigma_{ab}^3 \sigma_{\alpha\beta}^3 + \sigma_{ab}^+ \sigma_{\alpha\beta}^- + \sigma_{ab}^- \sigma_{\alpha\beta}^+ \right] \quad (66)$$

where $\sigma^\pm = \frac{1}{2}(\sigma^x \pm i\sigma^y)$ are the raising and lowering operators. Notice that the amplitude for the spin flip terms is twice that for the Ising terms. Since the propagators are diagonal in the spin indices, diagrams (1-3) only involve the Ising component in the interaction.

Taking account of the vanishing diagrams, we may compute the first three diagrams as follows:

$$\begin{aligned} 1. \quad & \text{Diagram 1: A loop with a central vertex connected to two external lines. The loop is shaded.} \\ & = \sum_m \frac{J_{mm}}{2} (2\rho B) M_o \\ & = (\bar{J}_R + \bar{J}_L) B M_o \\ 2. \quad & \text{Diagram 2: A loop with a central vertex connected to two external lines. The loop is shaded.} \\ & = \frac{1}{2} \sum_{m,n} \frac{1}{4} J_{mm} J_{nn} (2\rho B)^2 \left(-\frac{\partial M_o}{\partial B} \right) \\ & = -\frac{1}{2} (\bar{J}_R + \bar{J}_L)^2 B^2 \left(\frac{\partial M_o}{\partial B} \right) \\ 3. \quad & \text{Diagram 3: A loop with a central vertex connected to two external lines. The loop is shaded.} \\ & = \frac{1}{2} \text{Diagram 3: A loop with a central vertex connected to two external lines. The loop is shaded.} = 0 \end{aligned}$$

where the vanishing of the local conduction electron susceptibility (59) causes the last term to vanish.

The sum of the first three terms in the “free energy” is then

$$\Delta F_{(1)} + \Delta F_{(2)} + \Delta F_{(3)} = \bar{J} B M_o - \frac{1}{2} (\bar{J} B)^2 \frac{\partial M_o}{\partial B}, \quad (68)$$

where $M_o = \tanh(\frac{B}{T})$ is the magnetization of the free impurity, and we have put $\bar{J}_R + \bar{J}_L = \bar{J}$. We recognize these terms as the leading order expansion of the Free energy of a spin in a Weiss field $B_{eff} = (1 - J)B$:

$$\begin{aligned} F^* &= -T \ln [2 \cosh(B/T(1 - \bar{J}))] \\ &= -T \ln [2 \cosh(B/T)] + \bar{J} B M_o - \frac{1}{2} (\bar{J} B)^2 \frac{\partial M_o}{\partial B} + \dots \end{aligned} \quad (69)$$

The Ising corrections to the energy are clearly independent of the voltage between the leads.

The last diagram in the interaction energy can be expanded as follows:

$$4. \quad \frac{1}{2} \text{Diagram 4: A loop with a central vertex connected to two external lines. The loop is shaded.} = \frac{1}{2} \left[\text{Diagram 4a: A loop with a central vertex connected to two external lines. The loop is shaded.} + \text{Diagram 4b: A loop with a central vertex connected to two external lines. The loop is shaded.} \right] \quad (70)$$

These expressions can be divided into Ising (non-spin-flip) and x-y (spin flip) components, $\Delta F_{(4)} = \Delta F_{(4)}^z + \Delta F_{(4)}^\pm$, where

$$\begin{aligned} \Delta F_{(4)}^z &= \frac{1}{2} \left[\text{Diagram 4a: A loop with a central vertex connected to two external lines. The loop is shaded.} + \text{Diagram 4b: A loop with a central vertex connected to two external lines. The loop is shaded.} \right] \\ &= \sum_{m,n,\sigma} \frac{J_{mn}^2}{4} \int \frac{d\omega}{2\pi} (\pi_{m\sigma}^R \pi_{n\sigma}^K + \pi_{m\sigma}^K \pi_{n\sigma}^A)_\omega \\ \Delta F_{(4)}^\pm &= \frac{1}{2} \left[\text{Diagram 4c: A loop with a central vertex connected to two external lines. The loop is shaded.} + \text{Diagram 4d: A loop with a central vertex connected to two external lines. The loop is shaded.} \right] \\ &= \frac{1}{2} \sum_{m,n,\sigma} J_{mn}^2 \int \frac{d\omega}{2\pi} (\pi_{m\sigma}^R \pi_{n,-\sigma}^K + \pi_{m\sigma}^K \pi_{n,-\sigma}^A)_\omega \end{aligned} \quad (71)$$

(The $\frac{1}{2}$ prefactor in the first equation has been cancelled by the sum over the spin of the conduction electron lines, and the factor of $(J_{mn})^2/4$ is derived from the square of the Ising scattering amplitude. In the second equation, the spin of the conduction electrons is set by the spin of the f-electrons, so the $\frac{1}{2}$ prefactor remains, however, the amplitude for two spin flips is $(J_{mn})^2$, without a factor of $1/4$.) Inserting the discrete expressions (60) and (64) we obtain

$$\begin{aligned} \Delta F_{(4)}^z &= \text{Re} \sum_{mn\sigma} \frac{J_{mn}^2}{16} \left\{ \frac{(1 - h_\sigma h_{nk})(h_\sigma - h_{mk'})}{\epsilon_k - \epsilon_{k'}} \right\}, \\ \Delta F_{(4)}^\pm &= \text{Re} \sum_{mn\sigma} \frac{J_{mn}^2}{8} \left\{ \frac{(1 - h_{-\sigma} h_{nk})(h_\sigma - h_{mk'})}{\epsilon_k - \epsilon_{k'} - 2\sigma B} \right\}. \end{aligned} \quad (72)$$

where we have denoted $h_{mk} \equiv h_m(\epsilon_k)$. Following earlier discussion, the field dependence of the band-electron energies has been dropped in these expressions.

As a simple check on these results, consider the zero-temperature, equilibrium limit of (72). In this limit, one can replace $h_m(\epsilon_{k'}) \rightarrow -\text{sgn} \epsilon_{k'}$, $h_\sigma = \sigma$. The first term vanishes and the second term becomes

$$\begin{aligned} \Delta F_{(4)}^\pm &= \text{Re} \sum_{m,n,\sigma} \frac{J_{mn}^2}{8} \left\{ \frac{(1 - \sigma \text{sgn}(\epsilon_k))(\sigma + \text{sgn}(\epsilon_{k'}))}{\epsilon_k - \epsilon_{k'} - 2\sigma B} \right\} \\ &= J^2 \sum_{\epsilon_k < 0, \epsilon_{k'} > 0} \frac{1}{\epsilon_k - \epsilon_{k'} - 2B} \end{aligned} \quad (73)$$

which is recognized as the second-order correction to the ground-state energy due to quantum spin fluctuations.

To proceed further, it is convenient to introduce the notation

$$\begin{aligned} S_{mn\sigma}(x, B) &= \text{Re} \left[2\pi i \ln \tilde{\Gamma}(x + 2\sigma B + \mu_m - \mu_n) \right. \\ &\quad \left. - (x + 2\sigma B + \mu_m - \mu_n) \ln \left(\frac{D}{2\pi T} \right) \right] \\ \tilde{\Gamma}(x) &= \Gamma \left(\frac{1}{2} + \frac{x}{2\pi i} \right) \end{aligned} \quad (74)$$

which has the property that

$$\text{Re}\pi_{m\sigma}^R(x - \mu_n) = \rho S'_{mn\sigma}(x, B)$$

where the prime denotes derivative w.r.t. x . With this notation, using (63) and (65) we obtain

$$\begin{aligned}\Delta F_{(4)}^{\pm} &= \sum_{m,n} \frac{\bar{J}_{mn}^2}{4} \int d\omega S'_{mn\sigma}(\omega, B) (1 - \sigma M_o h(\omega)) \\ \Delta F_{(4)}^z &= \sum_{m,n} \frac{\bar{J}_{mn}^2}{8} \int d\omega S'_{mn\sigma}(\omega, 0) (1 + \sigma M_o h(\omega))\end{aligned}\quad (75)$$

where we have replaced $\text{Re}h_{-\sigma} = -\sigma M_o$. Integrating this expression by parts, and using the fact that $S_{mn\sigma}(\pm D) = \mp D$ we obtain

$$\begin{aligned}\Delta F_{(4)}^{\pm} &= - \sum_{m,n,\sigma} \bar{J}_{mn}^2 \left[D - \sum_{\sigma} \sigma \frac{M_o}{4} \int d\omega h'(\omega) S_{mn\sigma}(\omega) \right] \\ \Delta F_{(4)}^z &= - \sum_{m,n,\sigma} \bar{J}_{mn}^2 \frac{D}{2}\end{aligned}$$

where the residual integral in $\Delta F_{(4)}^z$ vanishes under the spin summation. It is convenient to separate the logarithmic term away from the integral over $S_{mn\sigma}(\omega)$, writing

$$\begin{aligned}J_{mn}^2 \sum_{\sigma} \sigma \frac{M_o}{4} \int d\omega [-h'(\omega)] S_{mn\sigma}(\omega) \\ = -2J_{mn}^2 B M_o \left[\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, \frac{\mu_m - \mu_n}{T}\right) \right]\end{aligned}\quad (77)$$

where

$$\phi(b, v) = \int \frac{dx}{(2 \cosh \frac{x}{2})^2} \frac{1}{4b} \sum_{\sigma, \gamma=\pm 1} 2\pi i \sigma \ln \tilde{\Gamma}(x + \sigma b + \gamma v) \quad (78)$$

Combining the results (76), (69), (77) and (78) our final result for the change in the Free energy $\Delta F = \int \frac{dJ}{J} \langle H_I \rangle$ is then

$$\begin{aligned}\Delta F &= -T \ln [2 \cosh(B(1 - \bar{J})/T)] - F_{eqn} - \frac{3}{2} \bar{J}^2 D \\ &+ 4\bar{J}_{RL}^2 M_o B \left[\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, \frac{eV}{T}\right) \right] \\ &+ 2(\bar{J}_R^2 + \bar{J}_L^2) M_o B \left[\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, 0\right) \right] \\ &+ O(\bar{J}^3).\end{aligned}\quad (79)$$

Notice that the appearance of F_{eqn} on the right hand side of this expression does not rely on any assumption about the limiting value of the Free- energy or magnetization in the limit $\bar{J} \rightarrow 0$. The change in the magnetization is then given by

$$\begin{aligned}\Delta M &= \left((1 - \bar{J}) \tanh \left[\frac{(1 - \bar{J})B}{T} \right] - \tanh \left[\frac{B}{T} \right] \right) \\ &- 4\bar{J}_{RL}^2 \frac{\partial}{\partial B} \left[B M_o \left(\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, \frac{eV}{T}\right) \right) \right]\end{aligned}$$

$$\begin{aligned}&- 2(\bar{J}_R^2 + \bar{J}_L^2) \frac{\partial}{\partial B} \left[B M_o \left(\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, 0\right) \right) \right] \\ &+ O(\bar{J}^3).\end{aligned}\quad (80)$$

Again, the appearance of the equilibrium magnetization on the right-hand side of this expression has nothing to do with the value of the magnetization in the limit $\bar{r}J \rightarrow 0$. With our method, we are unable to comment on the magnetization in the limit $\bar{J} \rightarrow 0$. However, the above expression is very strongly suggestive that the limit $\bar{J} \rightarrow 0$ returns to the equilibrium expression for the magnetization, in which case,

$$\begin{aligned}M &= (1 - \bar{J}) \tanh \left[\frac{(1 - \bar{J})B}{T} \right] \\ &- 4\bar{J}_{RL}^2 \frac{\partial}{\partial B} \left[B M_o \left(\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, \frac{eV}{T}\right) \right) \right] \\ &- 2(\bar{J}_R^2 + \bar{J}_L^2) \frac{\partial}{\partial B} \left[B M_o \left(\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, 0\right) \right) \right] \\ (76) &+ O(\bar{J}^3).\end{aligned}\quad (81)$$

There are two special limits of the above result. In the limit $T \rightarrow 0$, we may take advantage of the asymptotic form

$$\begin{aligned}B \left[\ln \frac{D}{2\pi T} - \phi\left(\frac{2B}{T}, \frac{V}{T}\right) \right] \longrightarrow \\ - \frac{1}{2} \sum_{\gamma=\pm} (2B + \gamma V) \ln \left[\frac{|2B + \gamma V|}{2\pi D e} \right]\end{aligned}\quad (82)$$

so that in this limit

$$\begin{aligned}M &= \left[1 - (\bar{J}_R + \bar{J}_L) - 2(\bar{J}_R^2 + \bar{J}_L^2) \ln \left(\frac{D}{2B} \right) \right. \\ &- \left. (4\bar{J}_{RL}^2) \ln \left(\frac{D}{\sqrt{[(eV)^2 - (2B)^2]}} \right) \right] \dots\end{aligned}\quad (83)$$

In the zero field limit $B \rightarrow 0$, $\phi(b, v) \rightarrow \phi(0, v) = \Phi(v) - 1 + \gamma$, where

$$\Phi(v) = \text{Re} \int_{-\infty}^{\infty} \frac{dx}{4 \cosh^2(\frac{x}{2})} \left[\tilde{\psi}(v+x) - \tilde{\psi}(x) \right] \quad (84)$$

and $\tilde{\psi}(x) = \psi(\frac{1}{2} + i\frac{x}{2\pi})$. and we have used the result

$$\int_{-\infty}^{\infty} \frac{dx}{4 \cosh^2(\frac{x}{2})} \tilde{\psi}(x) = -(1 + \gamma)$$

where $\gamma = 0.5772 \dots = -\psi(1)$ is the Euler constant. In this limit, the magnetization takes the form

$$\begin{aligned}M &= \frac{B}{T} \left\{ 1 - 2(\bar{J}_R + \bar{J}_L) - 4(\bar{J}_L^2 + \bar{J}_R^2) \ln \left(\frac{D e^{\frac{3}{4} + \gamma}}{2\pi T} \right) \right. \\ &- \left. 8|\bar{J}_{LR}|^2 \left[\ln \left(\frac{D e^{\frac{3}{4} + \gamma}}{2\pi T} \right) - \Phi\left(\frac{V}{T}\right) \right] \right\}.\end{aligned}\quad (85)$$

VIII. DISCUSSION

We should like to end with a discussion of some of the more controversial aspects of our work. There are two general topics that deserve discussion:

- * Does the magnetization of the quantum dot revert smoothly to the equilibrium value $M_o = \tanh\left(\frac{B}{T}\right)$ when the coupling to the leads is reduced to zero?
- * The issue of quantum coherence, and whether a current through the quantum dot dephases the Kondo effect.

Both issues are areas where our current work differs in important ways with parallel work carried out by others in the field.

1. Does the magnetization have a perturbative expansion?

The first of the above items brings our discussion into contact up with the issue of whether a perturbative treatment of the magnetization in the quantum dot is valid out-of-equilibrium. Although strictly speaking, our approach, based on the Onsager reciprocity relations, can not derive the leading term in the magnetization, the appearance of a well-defined perturbation expansion, and the very form of our results

$$\Delta M = \tanh(B(1 - \bar{J})/T) - \tanh(B/T) + O(\bar{J}^2) \quad (86)$$

suggests that the leading order term in the magnetization is just the Brillouin function $\tanh(B/T)$.

Recent work by Parcollet and Hooley (PH)²⁰, Rosch, Passke, Kroha and Wolfe³² and unpublished work by Kaminski and Glazman (KG)³⁴ casts doubt on the viability of a perturbative approach. PH compute the local magnetization of a quantum dot by computing the steady-state Green function of the local moment, and they conclude that in the limit where the coupling of the local moment to the leads becomes infinitesimal, the magnetization does not revert to its thermal value, but instead acquires the limiting value

$$\begin{aligned} \tilde{M}_{PH}(T, B, V) &= M_o f\left(\frac{2B}{T}, \frac{V}{T}\right) \\ f(b, v) &= \frac{\varphi(b)(1 + \theta)}{\frac{1}{2}(\varphi(b+v) + \varphi(b-v)) + \theta\varphi(b)} \end{aligned} \quad (87)$$

where

$$\varphi(x) = \frac{x}{\tanh \frac{x}{2}}$$

and $\theta = \frac{J_R^2 + J_L^2}{2J_{RL}^2}$. A similar expression was derived by KG using a master equation approach. Results in the recent paper by Rosch et al. also reduce to the same form. This expression can not be expanded perturbatively in

the coupling constants, and if correct, casts serious doubt in the whole viability of a perturbative approach to the non-equilibrium Kondo model.

The KGPH result assumes that the relevant magnetization of the quantum dot is the polarization of the local dot spin, whereas we have argued that because the spin of the quantum dot partially delocalizes into the leads, one must consider the global magnetization. This leads to a fundamentally different philosophy about the way a quantum dot equilibrates its magnetization. Clearly, the local magnetization is not a conserved quantity, and it can be relaxed by its coupling to the leads. By examining this problem, PH find a non-thermal polarization of the magnetic moment of the dot in the limit of zero coupling to the leads. We argue conversely, that if we focus on the *total magnetization* then the coupling to the leads is not sufficient to relax to the appropriate steady state magnetization. In this case, a weak coupling between the system and its surroundings is always required to relax the conserved total magnetization, which is otherwise conserved, even when the dot and the leads are coupled. Let us call the coupling between the dot and its surroundings s . It is reasonable to suppose that physically reasonable results will be obtained so long as s is small compared with all physical energy scales in the quantum dot, such as the renormalized Kondo temperature, but large compared with the adiabatic scale δ which governs the rate at which interactions are turned on:

$$\text{Physical energy scales, } (T_K^*, eV, \dots) \gg s \gg \delta$$

In KGPH approach, in the limit of zero coupling to the leads, the local moment of the quantum dot is left in a non-thermal polarization. These results are most striking in the limit of zero temperature, when the polarization of the quantum dot is given by

$$\tilde{M}_{PH} = \mu_B \times \begin{cases} \frac{2B(1+\theta)}{eV/\mu_B + 2\theta B} & (\mu_B B < eV/2) \\ 1 & (\mu_B B > eV/2) \end{cases}$$

for $k_B T \ll 2\mu_B B \ll eV$, where we have restored $V \rightarrow eV$ and $B \rightarrow \mu_B B$. To preserve this non-thermal distribution, the spin must remain decoupled from any other thermal bath. In other words, the PH result corresponds to the case where the coupling to an external thermal bath is zero, i.e $s = 0$.

To get a feeling for the consequences of this result, consider a quantum dot at a bias voltage of 100mV with $\theta = 1$. According to the PH result, the dot would have a magnetization $M \sim 0.1\mu_B$ even in a field of 50 Tesla. In a real experiment, -this magnetization would relax back to one Bohr magneton, due to coupling to an external bath. In practice, such a coupling is always present, and can not be neglected when considering the expansion around the limit $\bar{J} = 0$. Just as in equilibrium, providing these couplings are small compared with the relevant energy scales of the system- in this case the renormalized Kondo temperature- then a dynamical calculation of the magnetization must include them.

It is difficult to reconcile the proposed failure of a perturbative expansion for the magnetization out-of-equilibrium with the existence of a perturbative expansion for the current and the total interaction energy of the dot. The current and interaction energy of the quantum dot appear to have a perfectly regular perturbation expansion. In the steady state, then the magnetization is determined from differential of the coupling constant integrated interaction energy:

$$\Delta M = -\frac{\partial}{\partial B} \int_0^J \frac{dJ'}{J'} \langle H_I \rangle. \quad (88)$$

While the Onsager reciprocity relation does not hold for charge current and interaction energy, we are able to relate the current to the derivative of the interaction energy with respect to the interaction energy²⁶,

$$I = -\frac{\partial}{\partial A} \bigg|_{A=0} \langle H_I \rangle. \quad (89)$$

where the above quantities are to be calculated by including a vector potential A into the scattering potential (but not into the measurement vertex of H_I), by replacing

$$J_{RL} \rightarrow J_{RL} e^{i \frac{eA}{\hbar}}, \quad J_{LR} \rightarrow J_{LR} e^{-i \frac{eA}{\hbar}}.$$

Indeed, when we include these changes into our calculation of H_I , we do recover the established second order expression for the current through the quantum dot (Appendix D)

$$I = \frac{3e^2}{h}(\bar{J}_{RL})^2 V.$$

Thus a single energy functional appears to successfully yield both the magnetization and the current. From this point-of-view, a fundamental failure of perturbation theory would have to manifest itself in the interaction energy. The smooth perturbative expansion of this quantity again suggests that the problems encountered by PH in their calculation are one of equilibration, rather than a break down of the perturbation theory at high temperatures or fields.

From the arguments of this section, it appears that the proposed failure of the perturbative expansion is tied up with two issues- the appropriate definition of the Kondo magnetization, and the the issue of how the magnetization relaxes. If indeed, as we have argued, the total magnetization is the important variable for characterizing the Kondo effect, then a coupling to an external bath is necessary in any dynamic treatment of the total magnetization. It would be interesting in further work to examine whether such a coupling does dynamically relax the total magnetization to the energetically defined magnetization \mathcal{M} .

2. Decoherence and Ward Identities

Let us now turn to the second issue: that of decoherence. The general issue, whether a non-equilibrium quantum system loses its phase coherence is of great future importance, and will have bearing on non-equilibrium problems such as quantum computation. An issue here, fundamentally, is the question of whether driven non-equilibrium quantum systems can develop phase coherent structure, or not.

In our quantum dot problem, we have used the Onsager reciprocity relations to argue that the energetically defined magnetization

$$\mathcal{M} = M_{eqn} - \frac{\partial}{\partial B} \int_0^J \frac{dJ'}{J'} \langle H_I \rangle$$

is identically equal to the steady state magnetization of the quantum dot,

$$\mathcal{M} \equiv M \quad (90)$$

We are able to clearly demonstrate a perturbative expansion for \mathcal{M} . Clearly even if the identity between the energetic and the physical magnetization were to fail, the perturbative expansion of \mathcal{M} can still be used as a criterion for whether the Kondo effect enters a strong coupling regime at high voltages. The zero temperature, low field result

$$\mathcal{M} = \left[1 - J\rho - (J\rho)^2 \ln \left(\frac{D^2}{2B|(eV)|} \right) \dots \right] \quad (91)$$

shows that the *leading* Kondo logs associated with intralead processes survive to arbitrarily high voltage.

If decoherence is to prevent this divergence, at large voltages, then presumably, the above logs in \mathcal{M} would have to be cut-off by the decoherence rate $\tau^{-1} \sim I/e$. This would require that inside the logarithms, we replace

$$2B \rightarrow \max(2B, \hbar\tau^{-1}).$$

For weak coupling to the leads, $\hbar\tau^{-1} \sim \rho J^2 eV$ is the perturbative expansion for the current through the dot. Such cut-off effects will clearly appear if we extend the diagrammatics by dressing the fermion propagators using the Dyson equation

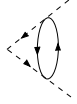
$$\tilde{\mathbf{G}}^{-1}(\omega) = \mathbf{G}_0^{-1}(\omega) - \text{---}\langle\!\!\langle \text{---} \rangle\!\!\rangle \quad (92)$$

The main effect of the self-energy insertions into the fermion lines is to introduce a wavefunction renormalization of the magnetization spin vertex

$$(Z-1)g\mu_B = \left\langle \frac{\partial \Sigma(\omega)}{\partial \omega} \right\rangle_{\omega=0} g\mu_B \quad (93)$$

However, in any controlled systematic treatment of the diagrammatics, it is important to also take into account

the corresponding renormalization of the magnetization vertex

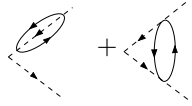


$$= \Gamma(\omega = 0)g\mu_B \quad (94)$$

Both of these diagrams are generated at the same time when one differentiates the interaction energy with respect to the external magnetic field. Now the important point is that these two vertices are related by a Ward identity associated with the conservation of total spin. In equilibrium, this leads to a Ward identity between the spin self-energy and the spin vertex

$$\Gamma_{\omega=0} = -\left.\frac{\partial \Sigma(\omega)}{\partial \omega}\right|_{\omega=0}$$

leading to the precise cancellation of these two terms.



$$= g\mu_B \left(\frac{\partial \Sigma(\omega)}{\partial \omega} + \Gamma \right)_{\omega=0} = 0 \quad (95)$$

This cancellation has important physical consequences. For example, this is the reason why the renormalized “meso-spin” $\vec{S}^*(\Lambda)$ of the sort discussed in the introduction

$$\vec{S}^* = e^{\vec{S}} \vec{S} e^{-\vec{S}}.$$

preserves a zero relaxation rate, whereas the bare spin \vec{S} relaxes at a rate $\tau^{-1} \sim J^2 k_B T$ at high temperatures. The point is, that the magnetization of the local spin M_{local} is rigorously given by

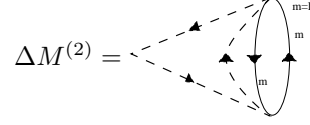
$$M_{local} = n_{\uparrow} - n_{\downarrow}$$

and this quantity therefore does not contain any vertex corrections. By contrast, the total magnetization entering into the uniform susceptibility of a quantum dot does contain the above vertex. From this we learn that the rapid fluctuations of the local spin \vec{S} described by the self-energy Σ are also responsible for the renormalization of the meso-spin $\vec{S}^*(\Lambda)$ and its delocalization into the leads. This is why these two effects cancel. These considerations lead us to doubt any conclusions about the behavior of the D.C. biased quantum dot which resum the self-energy corrections to the pseudo-fermion lines without taking the corresponding vertex corrections into account. Any calculations based on the “non-crossing approximation” which assume that these vertices can be ignored will lose these subtle cancellations.

It is useful to consider what will happen to the leading logarithm in the magnetic susceptibility if we continue to fourth order in the computation. The divergence we have found

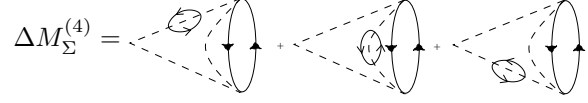
$$\Delta\chi = -\frac{4(\bar{J}_R^2 + \bar{J}_L^2)}{T} \ln \left(\frac{De^{1+\gamma}}{2\pi T} \right) \quad (96)$$

corresponds to the diagram



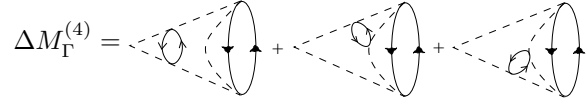
$$\Delta M^{(2)} = \quad (97)$$

where the logarithm derives from the f-conduction loops. When we go to fourth order, we dress the internal f-lines to produce the following three diagrams



$$\Delta M_{\Sigma}^{(4)} = \quad (98)$$

The renormalization effects of this diagram will tend to cut-off the logarithmic divergence. However to the same order we also produce vertex counterterms



$$\Delta M_{\Gamma}^{(4)} = \quad (99)$$

These vertex corrections correspond physically to the fact that the meso-spin $\vec{S}^*(\Lambda)$ is now partially delocalized into the leads, with the rapid fluctuations of the original spin \vec{S} now featuring as internal degrees of freedom inside the largely static meso-spin $\vec{S}^*(\Lambda)$. It is our contention that the Ward identity associated with spin conservation will cause the decoherence effects in $\Delta M_{\Sigma}^{(4)}$ to be cancelled by the vertex renormalizations in $\Delta M_{\Gamma}^{(4)}$, i.e.

$$\Delta M_{\Sigma}^{(4)} + \Delta M_{\Gamma}^{(4)} = 0,$$

and that furthermore, these cancellations will continue order by order. This is clearly an important point to confirm in future work.

We would like to end by discussing the nature of the low temperature phase that develops in a symmetric quantum dot, where $J_{RR} = J_{LL}$. In our previous work, we suggested that the intersite coupling terms J_{RL} are marginally irrelevant at large voltage bias, so that the underlying low temperature physics of the large bias quantum dot is that of a two channel Kondo model. In such a picture, the magnetic susceptibility would have a logarithmic divergence of the form

$$\chi \propto \frac{1}{T_K^*} \ln \left(\frac{T_K^*}{T} \right) \quad (100)$$

where $T_K^* \sim T_K^2/(eV)$. More recently, Rosch et al¹⁶ have suggested that the entry into the two channel Kondo regime will be interrupted at a temperature given by the decoherence scale

$$T_d \sim \frac{V}{\ln^2 \left(\frac{T_K}{T_d} \right)}$$

In the former picture, a Curie-type magnetic susceptibility would continue all the way down to temperatures of order T_K^* , whereas in the latter picture, the magnetic susceptibility would presumably saturate at a much higher temperature.

The survival of a Curie susceptibility at temperatures $T \ll T_d$ clearly relies on the existence of a renormalized composite “meso-spin” $S^*(\Lambda)$ and the vertex cancellation effects mentioned above. These same cancellation effects are intimately related to the ability of the meso-spin to transmit charge via internal fluctuations of the bare spin \vec{S} , without the meso-spin undergoing fluctuations at all. A simple physical picture of this phenomenon involves equal but opposite currents of electrons and holes to flowing with the meso-spin.

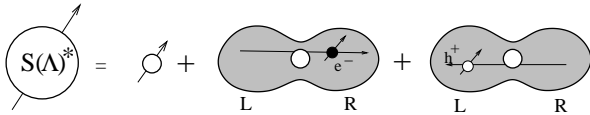


FIG. 7. Physical picture of how the “meso-spin” $S^*(\Lambda)$ can carry charge from the left lead to the right lead, through internal motion of spin within its composite structure. When the dot-spin migrates into the leads, a charge current can flow without a spin current if the current of holes balances the current of electrons.

At any given temperature, the original localized spin would then be in a state of rapid fluctuation, but the renormalized spin would remain essentially static until temperatures comparable with T_K^* .

3. Conclusion

This paper has discussed the nature of the magnetization in the DC biased Kondo effect, highlighting it as an important central element in the debate about whether the DC biased quantum dot is decohered by a finite current, or whether it is able to enter a new highly non-equilibrium strong coupling state. We have proposed a slightly radical idea that the principle of virtual work can be used in the non-equilibrium steady state by considering a large dot-lead system that is completely isolated, an idea that we showed is intimately related to our quantum reciprocity conjecture- the idea that subclass of variables in the non-equilibrium steady state continue to satisfy a reciprocity relation in time. Using these ideas, we computed an energetically defined magnetization and showed that it has a well-defined perturbation expansion in which the intra-lead Kondo logarithms are not cut-off by any finite voltage. This result supports our earlier work, based on a point-splitting perturbation procedures.

In the final discussion, we have attempted to bring out the most controversial elements in the ongoing debate. We have argued that most of the proposed differences with our work stem ultimately from a failure to consider the total magnetization of the Kondo problem, rather

than the local magnetization of the dot-spin. The total magnetization is not relaxed by coupling to the leads, and the physical susceptibility of real quantum dot always depends on a small coupling s to an external heat-bath. These difficulties can be avoided using an energetically defined definition of the magnetization, but they must be addressed directly if a dynamical calculation of the total magnetization is to be carried out. Finally, we have proposed that so-called de-coherence effects arise because of an incomplete and non-conserving treatment of the perturbation theory. When spin conservation is taken into account, cancellations take place between the wavefunction and vertex renormalization diagrams of the magnetization operator that ultimately are associated with the delocalization of the dot-spin into the leads. We have proposed that these cancellations continue into the non-equilibrium state leading to a complete preservation of quantum coherence in the strongly biased quantum dot. These issues will clearly be the subject of future work.

Experimentally, it is not yet feasible to measure the magnetization of individual quantum dots, however future improvements in the power of scanning Hall probes may make this a feasible experiment in the future. Another approach to the experimental examination of these phenomenon may lie in a new configuration. Magnetic atoms inside a wire between two electron baths at different voltages are exposed to two Fermi surface discontinuities, and their low temperature physics is likely to be very closely associated with the D.C. biased quantum dot.³³ It may be possible by measuring the magnetic susceptibility of pure silver wires containing rare-earth atoms with large Kondo temperatures, such as Cerium, to actually follow the magnetic aspects of the non-equilibrium Kondo problem. Experimental efforts in this direction would make an important counterparts to the theoretical debate we have outlined above.

Acknowledgments.

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IX. APPENDIX A: GREEN FUNCTIONS IN THE LARKIN OVCHINIKOV BASIS

We begin by writing the Green function in the Larkin Ovchinnikov Basis as

$$\mathbf{G}_{\alpha\beta}(1, 2) = -i\langle P\Psi_\alpha(1)\bar{\Psi}_\beta(2) \rangle$$

where

$$\Psi(1) \equiv (\psi_S(1), \psi_A(1)) \quad (101)$$

and

$$\bar{\Psi}(1) \equiv \Psi^\dagger(1)\tau_1 = \begin{pmatrix} \psi_A^\dagger(1) \\ \psi_S^\dagger(1) \end{pmatrix}. \quad (102)$$

Expanding this out gives

$$\mathbf{G} = -i \begin{pmatrix} \langle P\psi_S(1)\psi_A^\dagger(2) \rangle & \langle P\psi_S(1)\psi_S^\dagger(2) \rangle \\ \langle P\psi_A(1)\psi_A^\dagger(2) \rangle & \langle P\psi_A(1)\psi_S^\dagger(2) \rangle \end{pmatrix} \quad (103)$$

To transform this expression to its standard form, we need to expand the path-ordered Green-functions. Let us first carry this out for the $\langle P\psi_S(1)\psi_S^\dagger(2) \rangle$ term. Writing

$$\begin{aligned} \psi_S(1) &= \frac{1}{\sqrt{2}}(\psi(1_+) + \psi(1_-)) \\ \psi_A(1) &= \frac{1}{\sqrt{2}}(\psi(1_+) - \psi(1_-)) \end{aligned} \quad (104)$$

and then expanding the path-ordering along the Keldysh contour, we then have

$$\begin{aligned} -i\langle P\psi_S(1)\psi_S^\dagger(2) \rangle &= -\frac{i}{2} [\langle P\psi(1_+)\psi^\dagger(2_+) \rangle \\ &+ \langle P\psi(1_-)\psi^\dagger(2_-) \rangle + \langle P\psi(1_-)\psi^\dagger(2_+) \rangle + \langle P\psi(1_+)\psi^\dagger(2_-) \rangle] \\ &= -\frac{i}{2} [\langle T\psi(1)\psi^\dagger(2) \rangle + \langle \tilde{T}\psi(1)\psi^\dagger(2) \rangle + \langle [\psi(1), \psi^\dagger(2)] \rangle] \\ &= -i\langle [\psi(1), \psi^\dagger(2)] \rangle \equiv G_K(1, 2) \end{aligned}$$

in a similar fashion, we see the term $\langle P\psi_A(1)\psi_A^\dagger(2) \rangle$ term vanishes:

$$\begin{aligned} -i\langle P\psi_A(1)\psi_A^\dagger(2) \rangle &= -\frac{i}{2} [\langle P\psi(1_+)\psi^\dagger(2_+) \rangle \\ &+ \langle P\psi(1_-)\psi^\dagger(2_-) \rangle - \langle P\psi(1_-)\psi^\dagger(2_+) \rangle - \langle P\psi(1_+)\psi^\dagger(2_-) \rangle] \\ &= -\frac{i}{2} [\langle T\psi(1)\psi^\dagger(2) \rangle + \langle \tilde{T}\psi(1)\psi^\dagger(2) \rangle - \langle [\psi(1), \psi^\dagger(2)] \rangle] \\ &= 0 \end{aligned}$$

By contrast, the diagonal terms are given by

$$\begin{aligned} -i\langle P\psi_S(1)\psi_A^\dagger(2) \rangle &= -\frac{i}{2} [\langle P\psi(1_+)\psi^\dagger(2_+) \rangle \\ &- \langle P\psi(1_-)\psi^\dagger(2_-) \rangle + \langle P\psi(1_-)\psi^\dagger(2_+) \rangle - \langle P\psi(1_+)\psi^\dagger(2_-) \rangle] \\ &= -\frac{i}{2} [\langle T\psi(1)\psi^\dagger(2) \rangle - \langle \tilde{T}\psi(1)\psi^\dagger(2) \rangle + \langle [\psi(1), \psi^\dagger(2)] \rangle] \\ &= -i\langle \{\psi(1), \psi^\dagger(2)\} \rangle \theta(t_1 - t_2) \\ &\equiv G_R(1, 2) \end{aligned}$$

and

$$\begin{aligned} -i\langle P\psi_A(1)\psi_S^\dagger(2) \rangle &= -\frac{i}{2} [\langle P\psi(1_+)\psi^\dagger(2_+) \rangle \\ &+ \langle P\psi(1_-)\psi^\dagger(2_-) \rangle + \langle P\psi(1_-)\psi^\dagger(2_+) \rangle + \langle P\psi(1_+)\psi^\dagger(2_-) \rangle] \\ &= -\frac{i}{2} [\langle T\psi(1)\psi^\dagger(2) \rangle - \langle \tilde{T}\psi(1)\psi^\dagger(2) \rangle - \langle [\psi(1), \psi^\dagger(2)] \rangle] \\ &= +i\langle \{\psi(1), \psi^\dagger(2)\} \rangle \theta(t_2 - t_1) \\ &\equiv G_A(1, 2) \end{aligned}$$

With these manipulations, we see that we can write

$$\mathbf{G}(1, 2) = \begin{pmatrix} G_R(1, 2) & G_K(1, 2) \\ 0 & G_A(1, 2) \end{pmatrix}$$

X. APPENDIX B: FEYNMAN RULES FOR INTERACTION VERTICES

To develop the Feynman rules for perturbation theory, we need to know the matrix elements of the interaction H_I the Larkin Ovchinnikov basis. To compute these, let us consider the simplest case of an interaction

$$H_I = \frac{1}{2} \int d1 d2 V(1-2) \rho(1) \rho(2). \quad (105)$$

where $\rho(1) = \psi^\dagger(1)\psi(1)$ is the density. We can combine the density operator on the upper and lower contours of the Keldysh path to make a symmetric and antisymmetric combination

$$\begin{aligned} \rho_S(1) &= \frac{1}{2}(\rho(1_+) + \rho(1_-)) \\ \rho_A(1) &= (\rho(1_+) - \rho(1_-)) \end{aligned}$$

The first combination is the measured value of the particle number, whereas the second combination is the field that couples to an external chemical potential. In the Larkin Ovchinnikov basis, these two operators can be written

$$\begin{aligned} \rho_S(1) &= \frac{1}{2}(\psi_S^\dagger(1)\psi_S(1) + \psi_A^\dagger(1)\psi_A(1)) = \frac{1}{2}\bar{\Psi}(1)\tau_1\Psi(1) \\ \rho_A(1) &= (\psi_S^\dagger(1)\psi_A(1) + \psi_A^\dagger(1)\psi_S(1)) = \bar{\Psi}(1)\Psi(1) \end{aligned}$$

so that the “measurement vertex”, $\tau_1/2$ is off-diagonal whereas the response vertex is diagonal. This Keldysh structure holds for any one-particle operator.

On the Keldysh contour we can define the measurement and response fields for H_I as follows

$$\begin{aligned} H_I^S &= \frac{1}{4} \int d1 d2 V(1-2) [\rho(1_+)\rho(2_+) + \rho(1_-)\rho(2_-)] \\ H_I^A &= \frac{1}{2} \int d1 d2 V(1-2) [\rho(1_+)\rho(2_+) - \rho(1_-)\rho(2_-)]. \end{aligned} \quad (106)$$

The first of these operators is used to compute the expectation value of the interaction, but it is the second operator which enters into the time-ordered exponential

and provides the scattering vertices that make up the Feynman diagrams. These can be re-written as

$$\begin{aligned} H_I^S &= \frac{1}{2} \int d1 d2 V(1-2) [\rho^S(1) \rho^S(2) + \frac{1}{4} \rho^A(1) \rho^A(2)] \\ H_I^A &= \frac{1}{2} \int d1 d2 V(1-2) [\rho^S(1) \rho^A(2) + \rho^A(1) \rho^S(2)]. \end{aligned} \quad (107)$$

The second term in H^S involves the product of two antisymmetric operators, whose expectation value is zero. Since we only ever wish to calculate the expectation value of H_I , we can drop these terms. This enables us to write the interaction operators in the form

$$\begin{aligned} H_I^S &= \frac{1}{2} \int d1 d2 \tilde{V}_{\lambda\lambda',\gamma\gamma'}^S (1-2) \bar{\Psi}_\lambda(1) \bar{\Psi}_\gamma(2) \Psi_{\gamma'}(2) \Psi_{\lambda'}(1) \\ H_I^A &= \frac{1}{2} \int d1 d2 \tilde{V}_{\lambda\lambda',\gamma\gamma'}^A (1-2) \bar{\Psi}_\lambda(1) \bar{\Psi}_\gamma(2) \Psi_{\gamma'}(2) \Psi_{\lambda'}(1) \end{aligned} \quad (108)$$

where the Keldysh matrix elements for the measurement and scattering vertices are

$$\begin{aligned} \tilde{V}_{\lambda\lambda',\gamma\gamma'}^S (1-2) &= V(1-2) \left[\frac{\tau_1}{2} \otimes \frac{\tau_1}{2} \right]_{\lambda\lambda',\gamma\gamma'} \\ \tilde{V}_{\lambda\lambda',\gamma\gamma'}^A (1-2) &= V(1-2) \left[\frac{\tau_1}{2} \otimes \mathbb{1} + \mathbb{1} \otimes \frac{\tau_1}{2} \right]_{\lambda\lambda',\gamma\gamma'} \end{aligned} \quad (109)$$

In a Feynman diagram, each of these vertices is associated with two propagators, introducing a factor (i^2). The antisymmetric vertex also picks up a factor of $-i$ derived from the path ordered exponential. The final results for the scattering vertices in the Feynman diagrams is then

$$\begin{aligned} V_{\lambda\lambda',\gamma\gamma'}^S (1-2) &= V(1-2) \left[\frac{i\tau_1}{2} \otimes \frac{i\tau_1}{2} \right]_{\lambda\lambda',\gamma\gamma'} \\ V_{\lambda\lambda',\gamma\gamma'}^A (1-2) &= V(1-2) \left[\frac{i\tau_1}{2} \otimes \mathbb{1} + \mathbb{1} \otimes \frac{i\tau_1}{2} \right]_{\lambda\lambda',\gamma\gamma'} \end{aligned} \quad (110)$$

These results are quickly generalized to spin dependent interactions. In this paper, we are interested in the Kondo interactions of the form

$$H_I = \frac{J}{2} f^\dagger_\alpha \vec{\sigma}_{\alpha\beta} f_\beta \psi_a \vec{\sigma}_{ab} \psi_b \quad (111)$$

so the interaction vertex is modified by replacing

$$V(1-2) \rightarrow \frac{J}{2} \vec{\sigma}_{ab} \cdot \vec{\sigma}_{\alpha\beta}$$

The corresponding interaction vertices are then

$$\begin{aligned} V^S &\equiv \frac{J}{2} \vec{\sigma}_{ab} \cdot \vec{\sigma}_{\alpha\beta} \left[\frac{i\tau_1}{2} \otimes \frac{i\tau_1}{2} \right]_{\lambda\lambda',\gamma\gamma'} \\ V^A &\equiv \frac{J}{2} \vec{\sigma}_{ab} \cdot \vec{\sigma}_{\alpha\beta} \left[\frac{i\tau_1}{2} \otimes \mathbb{1} + \mathbb{1} \otimes \frac{i\tau_1}{2} \right]_{\lambda\lambda',\gamma\gamma'} \end{aligned} \quad (112)$$

where for clarity, we have omitted the indices from V^S and V^A .

XI. APPENDIX C: DERIVATION OF INTEGRAL IDENTITY

In this section we prove two useful results. Firstly, that

$$I_0 = \int \frac{d\omega}{\pi} \left[f(\omega) - \frac{1}{2} \right] \left[\frac{1}{\omega - \zeta} - \frac{1}{\omega - \zeta'} \right] \quad (113)$$

$$= \frac{1}{\pi} \left[\psi \left(\frac{1}{2} + \frac{\zeta}{2\pi iT} \right) - \psi \left(\frac{1}{2} + \frac{\zeta'}{2\pi iT} \right) \right]. \quad (114)$$

where $\psi(z) = d \ln \Gamma(z) / dz$ is the digamma function and second that

$$\begin{aligned} I_1 &= \int \frac{d\omega}{2\pi} f(\omega) \Phi(\omega) \frac{1}{\omega - \zeta} \\ &= \frac{1}{\pi} \left[\psi \left(\frac{1}{2} + \frac{\zeta}{2\pi iT} \right) - \ln \left(\frac{D}{2\pi iT} \right) \right]. \end{aligned} \quad (115)$$

where $\Phi(\omega) = D^2 / (\omega^2 + D^2)$ is a Lorentzian cut-off function.

Consider the first integral I_0 . To evaluate it, we let ζ and ζ' both lie in the upper half complex plane. By completing the integral around the series of poles in the function $f(z)$ at $z = -i\omega_n$, we obtain

$$\begin{aligned} I_0 &= 2iT \sum_{\omega_n < 0} \left(\frac{1}{i\omega_n - \zeta} - \frac{1}{i\omega_n - \zeta'} \right) \\ &= -\frac{1}{\pi} \sum_{n=1}^{\infty} \left(\frac{1}{n + \frac{1}{2} + \frac{\zeta}{2\pi iT}} - \frac{1}{n + \frac{1}{2} + \frac{\zeta'}{2\pi iT}} \right) \end{aligned} \quad (116)$$

But the digamma function can be expanded as a series,

$$\psi(z) = -C + \sum_{n=0}^{\infty} \left(\frac{1}{n+1} - \frac{1}{n+z} \right), \quad (117)$$

where $-C = \psi(1)$ is the Euler constant, so that

$$\psi(x) - \psi(y) = \sum_{n=0}^{\infty} \left(\frac{1}{n+y} - \frac{1}{n+x} \right). \quad (118)$$

Substituting this into (116), we then obtain

$$I_0 = \frac{1}{\pi} \left[\psi \left(\frac{1}{2} + \frac{\zeta}{2\pi iT} \right) - \psi \left(\frac{1}{2} + \frac{\zeta'}{2\pi iT} \right) \right] \quad (119)$$

Let us turn to the second integral

$$I_1 = \int \frac{d\omega}{\pi} \left[f(\omega) - \frac{1}{2} \right] \Phi(\omega) \frac{1}{\omega - \zeta}, \quad (120)$$

where $\Phi(\omega) = D^2 / (\omega^2 + D^2)$. Expanding out the cut-off function, writing

$$\Phi(\omega) = \frac{D}{2i} \left[\frac{1}{\omega - iD} - \frac{1}{\omega + iD} \right],$$

then

$$I_1 = \frac{D}{2i} \int \frac{d\omega}{\pi} \left[f(\omega) - \frac{1}{2} \right] \left[\frac{1}{\omega - \zeta} \frac{1}{\omega - iD} + \{D \rightarrow -D\} \right]$$

in the limit where $D \gg |\zeta|$, we have

$$I_1 = \int \frac{d\omega}{2\pi} \left[f(\omega) - \frac{1}{2} \right] \left[\left(\frac{1}{\omega - \zeta} - \frac{1}{\omega - iD} \right) + \{D \rightarrow -D\} \right].$$

But the second term is equal to the first one, because

$$\int \frac{d\omega}{2\pi} \left[f(\omega) - \frac{1}{2} \right] \frac{1}{\omega - iD} = \int \frac{d\omega}{2\pi} \left[f(\omega) - \frac{1}{2} \right] \frac{1}{\omega + iD}$$

(the difference between these two integrals is the integral of an odd, with an even function, so it vanishes), so that

$$I_1 = \int \frac{d\omega}{\pi} \left[f(\omega) - \frac{1}{2} \right] \left[\left(\frac{1}{\omega - \zeta} - \frac{1}{\omega - iD} \right) \right]. \quad (121)$$

Using Eq.(119), we get

$$\begin{aligned} & \int \frac{d\omega}{\pi} \left[f(\omega) - \frac{1}{2} \right] \Phi(\omega) \frac{1}{\omega - \zeta} \\ &= \frac{1}{\pi} \left[\psi\left(\frac{1}{2} + \frac{\zeta}{2\pi iT}\right) - \psi\left(\frac{1}{2} + \frac{D}{2\pi T}\right) \right] \\ &= \frac{1}{\pi} \left[\psi\left(\frac{1}{2} + \frac{\zeta}{2\pi iT}\right) - \ln\left(\frac{D}{2\pi T}\right) \right]. \end{aligned} \quad (122)$$

Finally, since

$$\int \frac{d\omega}{2\pi} \Phi(\omega) \frac{1}{\omega - \zeta} = \frac{i}{2}, \quad (123)$$

we reach

$$\int \frac{d\omega}{2\pi} f(\omega) \Phi(\omega) \frac{1}{\omega - \zeta} = \frac{1}{\pi} \left[\psi\left(\frac{1}{2} + \frac{\zeta}{2\pi iT}\right) - \ln\left(\frac{D}{2\pi iT}\right) \right]. \quad (124)$$

XII. APPENDIX D: DERIVATION OF CHARGE CURRENT AND CONDUCTANCE

In this section, we derive an expression for charge current to the second order of J using the interaction energy $\langle H_I \rangle$. While the Onsager reciprocity relation does not hold for charge current and interaction energy, we are able to relate the current to the derivative of the interaction energy with respect to the interaction energy²⁶,

$$I = -\frac{\partial}{\partial A} \Big|_{A=0} \langle H_I \rangle. \quad (125)$$

where the above quantities are to be calculated by including a vector potential A into the scattering potential (but not into the measurement vertex of H_I), by replacing

$$J_{RL} \rightarrow J_{RL} e^{-i\frac{eA}{\hbar}}, \quad J_{LR} \rightarrow J_{LR} e^{i\frac{eA}{\hbar}}.$$

After making this gauge transform we obtain the following expression for the interaction energy to second order (at zero magnetic field $B = 0$),

$$\begin{aligned} \langle H_I^{(2)} \rangle &= 3J_{LR}^2 \int \frac{d\epsilon}{2\pi} \\ &\left\{ \left[\pi_L^R(\epsilon) \pi_R^K(\epsilon) + \pi_L^K(\epsilon) \pi_R^A(\epsilon) \right] e^{-\frac{i\epsilon A}{\hbar}} \right. \\ &\quad \left. + \left[\pi_R^R(\epsilon) \pi_L^K(\epsilon) + \pi_R^K(\epsilon) \pi_L^A(\epsilon) \right] e^{\frac{i\epsilon A}{\hbar}} \right\} \end{aligned} \quad (126)$$

where the factor 3 comes from summation of spins, Eq.(66). By differentiating with respect to the vector potential, we obtain

$$\begin{aligned} I^{(2)} &= 3J_{LR}^2 \frac{ie}{\hbar} \int \frac{d\epsilon}{2\pi} \left\{ -\left[\pi_L^K(\epsilon) (\pi_R^R(\epsilon) - \pi_R^A(\epsilon)) \right] \right. \\ &\quad \left. + \left[\pi_R^K(\epsilon) (\pi_L^R(\epsilon) - \pi_L^A(\epsilon)) \right] \right\} \end{aligned} \quad (127)$$

By using Eq.(63) and (65), the zero temperature current is then

$$\begin{aligned} I^{(2)} &= 3J_{LR}^2 \frac{e\pi\rho^2}{\hbar} \int \frac{d\epsilon}{2\pi} \text{Im} \left[\psi\left(\frac{1}{2} + \frac{\epsilon + \mu_R}{2\pi iT}\right) - \psi\left(\frac{1}{2} + \frac{\epsilon + \mu_L}{2\pi iT}\right) \right] \\ &= 3J_{LR}^2 \frac{e^2}{\hbar} \pi\rho^2 V, \end{aligned} \quad (128)$$

hence

$$G = \frac{3e^2\pi}{\hbar} J_{LR}^2 \rho^2. \quad (129)$$

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- ¹ M. A. Kastner, Rev. Mod. Phys. **64**, 849 (1992); R. C. Ashoori, Nature **379**, 413 (1996); L. P. Kouwenhoven and C. Marcus, Physics World **11**, 35 (June 1998).
 - ² T. K. Ng and P. A. Lee, Phys. Rev. Lett. **61**, 1768 (1988).
 - ³ L. I. Glazman and M. E. Raikh, Pis'ma Zh. Eksp. Teor. Fiz. **47**, 378 (1988) [JETP Letters **47**, 452 (1988)].
 - ⁴ Y. Meir and N. S. Wingreen, Phys. Rev. Lett. **68**, 2512 (1992).
 - ⁵ Y. Meir, N. S. Wingreen, and P. A. Lee, Phys. Rev. Lett. **70**, 2601 (1993).
 - ⁶ N. Wingreen and Y. Meir, Phys. Rev. **B49**, 11040 (1994).
 - ⁷ D. Goldhaber-Gordon *et al.*, Nature **391**, 156 (1998).
 - ⁸ S. M. Cronenwett, T. H. Oosterkamp, and L. P. Kouwenhoven, Science **281**, 540 (1998).
 - ⁹ W. G. van der Wiel *et al.*, Science **289**, 2105 (2000).
 - ¹⁰ A. Kaminski, Yu. V. Nazarov, and L. I. Glazman, Phys. Rev. **B 62**, 8154(2000).
 - ¹¹ M. Plihal, D. C. Langreth, P. Nordlander, Phys. Rev. **B61**, R13341 (2000).
 - ¹² A. Schiller and S. Hershfield, Phys. Rev. **B58**, 14978 (1998).
 - ¹³ Y. Avishai and Y. Goldin, Phys. Rev. Lett. **81**, 5394 (1998).
 - ¹⁴ P. W. Anderson, Comm. Solid St. Phys. **5**, 72 (1973).

- ¹⁵ A. Kaminski, Yu. V. Nazarov, and L. I. Glazman, Phys. Rev. Lett. **83**, 384 (1999).
- ¹⁶ A. Rosch, J. Kroha and P. Wölfe, Phys. Rev. Lett. **87**, 156802 (2001).
- ¹⁷ A. Schiller and S. Hershfield, Phys. Rev. B **62**, R16271 (2000).
- ¹⁸ X.-G. Wen, LANL preprint `cond-mat/9812431`.
- ¹⁹ P. Coleman, C. Hooley and O. Parcollet, Phys. Rev. Lett. **86** 4088 (2001).
- ²⁰ O. Parcollet and C. Hooley, to be published `cond-mat/0202425`.
- ²¹ L. Onsager, Physical Review **37**, 405 (1931)
- ²² L. Onsager, Physical Review **38**, 2265 (1931).
- ²³ For a review of Keldysh method, see J. Rammer and H. Smith, Rev. Mod. Phys., **58**, 323 (1986).
- ²⁴ Lord Rayleigh, Proc. Math. Soc London **4**, 357, [363], (1873); Theory of Sound (London, MacMillan Co, 1st Ed 1877), Vol 1, p 78 (2d ed 1878), Vol 1, p 102.
- ²⁵ Larkin, A. I., and Yu.N. Ovchinnikov, 1975, Zh. Eksp. Teor. Fiz. **68**, 1915 (Sov. Phys.-JETP **41**, 960 (1975)).
- ²⁶ P. Coleman and W. Mao, to be published.
- ²⁷ B. Derrida, J. L. Lebowitz and E. R. Speer, Phys. Rev. Lett. **87**, 1506001 (2001)); see also L. Bertini et al., Phys. Rev. Lett. **87**, 040601 (2001).
- ²⁸ A.A. Abrikosov, Physics **2**, 5 (1965).
- ²⁹ V. N. Popov and S. A. Fedotov, Sov. Phys. JETP **67**, 535 (1988).
- ³⁰ M.N. Kiselev and R. Oppermann, Phys. Rev. Lett. **85**, 5631 (2000).
- ³¹ A. Kaminski and L. Glazman, private communication (2001).
- ³² A. Rosch, J. Passke, J. Kroha and P. Wölfe, `cond-mat/0202404` (2002).
- ³³ H. Pothier et al., Phys. Rev. Lett. **79**, 3490 (1997);
- ³⁴ A. Kaminskii and L. Glazman, Phys. Rev. Lett. **86**, 2400 (2001).